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Appendix D

Gamma Spectroscopy Screening

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This appendix provides the results of gamma spectroscopy ^{241}Am and $^{239,240}\text{Pu}$ compared with inductively coupled plasma-mass spectroscopy (ICP-MS) ^{239}Pu results for waste and soil samples. The radionuclides which would produce measurable values include ^{241}Am , ^{239}Pu , and ^{240}Pu . The gamma energies from the plutonium isotopes are very similar, and are not resolvable, and hence are reported as a single value, designated $^{239+240}\text{Pu}$. Results of the analyses are presented in Table D-1, together with subsequently generated ICP-MS results for ^{241}Am and ^{239}Pu , which were included to facilitate comparison. Isotopes other than ^{241}Am and ^{239}Pu identified in the gamma spectroscopy screen analysis are included in Table D-2.

Table D-1. Gamma spectroscopy and ICP-MS results for waste and soil samples for ^{241}Am , $^{239,240}\text{Pu}$ (gamma), and ^{239}Pu (ICP-MS). The ranges reported for the gamma spectroscopy measurements are standard deviations derived from counting statistics. The values reported for the ICP-MS measurements are averages and standard deviations from a minimum of three measurements.

Sample Number	Acquisition Date	Appearance Category	Gamma Spectroscopy ^{241}Am (nCi/g)	Gamma Spectroscopy $^{239+240}\text{Pu}$ (nCi/g)	ICP-MS $^{241}\text{Am}^*$ (nCi/g)	ICP-MS ^{239}Pu (nCi/g)
Fallout	n/a	—	0.03–0.0005	0.2–0.002	0.03–0.0005	0.2–0.002
Benchmark Samples						
BLANK SOIL	—	Clean soil	—	—	<0.52	<1.2
P9GW <u>04</u> 013A	—	Clean soil	—	—	<0.52	<1.2
P9GW <u>09</u> 013A	1/27/2004	Clean soil	—	—	<0.52	<1.2
P9GW <u>12</u> 013A	—	Clean soil	—	—	<0.52	<1.2
P9GW <u>13</u> 013A	1/27/2004	Clean soil	—	—	<0.52	<1.2
P9GW <u>15</u> 013A	1/29/2004	Clean soil	—	—	<0.52	<1.2
P9GW <u>21</u> 013A	—	Clean soil	—	—	<0.52	<1.2
Interstitial Sample - Clean Soil						
P9GT <u>09</u> 016G	2/13/2004	Clean soil	0.833 ± 0.076	<5.2	2.9 ± 3.9	2.8 ± 2.3
P9GT <u>10</u> 016G	2/14/2004	Clean soil	1.122 ± 0.067	<5.4	1.42 ± 0.53	3.4 ± 1.3
P9GT <u>13</u> 016G	2/17/2004	Clean soil	0.103 ± 0.017	<4.1	<0.52	<1.2
P9GT <u>21</u> 016G	2/17/2004	Clean soil	0.467 ± 0.032	<4.9	0.64 ± 0.33	1.43 ± 0.48
P9GT <u>22</u> 016G	2/17/2004	Clean soil	0.0087 ± 0.0019	<4.6	<0.52	<1.2
P9GT <u>24</u> 016G	2/17/2004	Clean soil	0.160 ± 0.017	<4.8	<0.52	<1.2
P9GT <u>28</u> 016G	2/19/2004	Clean soil	334 ± 27	$5,110 \pm 650$	450 ± 120	$2,170 \pm 560$
P9GT <u>32</u> 016G	2/19/2004	Clean soil	434 ± 37	<330	760 ± 360	$3,700 \pm 1,800$
P9GT <u>34</u> 016G	2/19/2004	Clean soil	451 ± 43	<690	790 ± 120	$3,770 \pm 660$
Interstitial Sample – Clean-to-mostly clean soil						
P9GT <u>08</u> 016G	2/13/2004	Clean-to-mostly clean soil	0.407 ± 0.038	<4.9	0.74 ± 0.35	3.8 ± 1.9
P9GT <u>11</u> 016G	2/14/2004	Clean-to-mostly clean soil	2.45 ± 0.30	9.8 ± 1.6	2.1 ± 1.1	4.62 ± 0.61
P9GT <u>12</u> 016G	2/14/2004	Clean-to-mostly clean soil	1.542 ± 0.094	<5.6	1.61 ± 0.77	5.3 ± 2.5
P9GT <u>14</u> 016G	2/17/2004	Clean-to-mostly clean soil	0.304 ± 0.028	<3.8	<0.52	1.67 ± 0.74
P9GT <u>15</u> 016G	2/17/2004	Clean-to-mostly clean soil	0.211 ± 0.023	<4.7	1.38 ± 0.31	21 ± 32
P9GT <u>16</u> 016G	2/17/2004	Clean-to-mostly	0.358 ± 0.022	<5.2	<0.52	1.28 ± 0.78

Table D-1. (continued).

Sample Number	Acquisition Date	Appearance Category	Gamma Spectroscopy ^{241}Am (nCi/g)	Gamma Spectroscopy $^{239+240}\text{Pu}$ (nCi/g)	ICP-MS $^{241}\text{Am}^*$ (nCi/g)	ICP-MS ^{239}Pu (nCi/g)
		clean soil				
P9GT <u>18</u> 016G	2/17/2004	Clean-to-mostly clean soil	0.124 ± 0.047	<4.5	<0.52	1.34 ± 0.19
P9GT <u>20</u> 016G	2/17/2004	Clean-to-mostly clean soil	1.038 ± 0.063	<3.75	<0.52	2.1 ± 1.4
P9GT <u>23</u> 016G	2/17/2004	Clean to mostly clean soil	0.489 ± 0.058	<4.7	0.54 ± 0.63	1.41 ± 0.64
P9GT <u>25</u> 016G	2/17/2004	Clean-to-mostly clean soil	0.302 ± 0.044	<5.4	0.73 ± 0.70	1.9 ± 1.7
P9GT <u>26</u> 016G	2/17/2004	Clean-to-mostly clean soil	0.206 ± 0.015	<5.5	<0.52	<1.2
P9GT <u>29</u> 016G	2/19/2004	Clean to mostly clean soil	380 ± 32	8,990 ± 850	370 ± 160	1,850 ± 850
P9GT <u>30</u> 016G	2/19/2004	Clean to mostly clean soil	412 ± 37	5,070 ± 600	960 ± 210	4,100 ± 1,500
P9GT <u>31</u> 016G	2/19/2004	Clean to mostly clean soil	432 ± 41	13,000 ± 1,000	530 ± 160	2,630 ± 850
P9GT <u>33</u> 016G	2/19/2004	Clean-to-mostly clean soil	396 ± 40	3,600 ± 400	830 ± 760	3,900 ± 3,700
P9GT <u>35</u> 016G	2/19/2004	Clean-to-mostly clean soil	389 ± 39	4,680 ± 620	680 ± 290	3,300 ± 1,500
P9GT <u>36</u> 016G	2/19/2004	Clean-to-mostly clean soil	414 ± 35	4,730 ± 430	760 ± 190	3,620 ± 830
Interstitial Sample – Mixed Soil-unknown waste						
P9GT <u>01</u> 016G	2/2/2004	Mixed soil-unknown waste	157 ± 12	<46	340 ± 480	16 ± 21
P9GT <u>02</u> 016G	2/2/2004	Mixed soil-unknown waste	448 ± 42	<500	880 ± 850	43 ± 41
P9GT <u>03</u> 016G	2/8/2004	Mixed soil-unknown waste	14.82 ± 0.93	<4.5	14 ± 11	2.3 ± 1.0
P9GT <u>04</u> 016G	2/8/2004	Mixed soil-unknown waste	1.36 ± 0.12	<5.6	1.23 ± 0.39	4.5 ± 3.2
P9GT <u>05</u> 016G	2/8/2004	Mixed soil-unknown waste	27.0 ± 1.7	<5.6	16 ± 12	1.29 ± 0.85
P9GT <u>06</u> 016G	2/8/2004	Mixed	1.16 ± 0.12	<5.0	1.9 ± 1.9	4.3 ± 2.0

Table D-1. (continued).

Sample Number	Acquisition Date	Appearance Category	Gamma Spectroscopy ^{241}Am (nCi/g)	Gamma Spectroscopy $^{239+240}\text{Pu}$ (nCi/g)	ICP-MS $^{241}\text{Am}^*$ (nCi/g)	ICP-MS ^{239}Pu (nCi/g)
		soil-unknown waste				
P9GT <u>07</u> 016G	2/12/2004	Mixed soil-unknown waste	0.919 ± 0.063	5 ± 13	2.57 ± 0.99	7.00 ± 0.99
P9GT <u>17</u> 016G	2/17/2004	Mixed soil-unknown waste	0.886 ± 0.083	<6.2	1.72 ± 0.69	4.2 ± 2.3
P9GT <u>19</u> 016G	2/17/2004	Mixed soil-unknown waste	1.20 ± 0.10	<6.3	1.86 ± 0.75	8.94 ± 0.51
P9GT <u>27</u> 016G	2/17/2004	Mixed soil-unknown waste	283 ± 20	<140	$1,000 \pm 1,100$	$4,900 \pm 5,900$
P9GT <u>27</u> 016G (duplicate ICP-MS)	2/17/2004	Mixed soil-unknown waste	283 ± 20	<140	471 ± 70	$1,929 \pm 66$
Organic Sample – Organic sludge probable						
P9GR <u>04</u> 012G	2/1/2004	Organic sludge probable	$2,420 \pm 200$	<840	$6,000 \pm 4,400$	300 ± 230
P9GR <u>04</u> 012G (duplicate ICP-MS)	2/1/2004	Organic sludge probable	$2,420 \pm 200$	<840	$3,480 \pm 260$	173 ± 28
P9GR <u>20</u> 012G	2/2/2004	Organic sludge probable	$1,540 \pm 130$	<320	$1,560 \pm 320$	76 ± 16
P9GR <u>23</u> 012G	2/12/2004	Organic sludge probable	4.27 ± 0.39	<8.76	12 ± 11	54 ± 56
Inorganic Sample – Unknown Waste						
P9GP <u>01</u> 015G	2/1/2004	Unknown waste material	2.36 ± 0.21	<15.2	1.8 ± 1.0	4.9 ± 8.3
P9GP <u>02</u> 015G	2/1/2004	Unknown waste material	$2,310 \pm 190$	$8,500 \pm 1,200$	$2,170 \pm 400$	102 ± 21
P9GP <u>03</u> 015G	2/2/2004	Unknown waste material	26.5 ± 2.4	<13.5	17 ± 19	5.7 ± 9.4
P9GP <u>04</u> 015G	2/2/2004	Unknown waste material	$2,210 \pm 180$	$4,760 \pm 440$	$1,010 \pm 760$	47 ± 64
P9GP <u>05</u> 015G	2/2/2004	Unknown waste material	$1,600 \pm 170$	<253	$1,100 \pm 120$	58.0 ± 7.3

Table D-2. Isotopes other than ^{241}Am and $^{239-240}\text{Pu}$ identified in the gamma spectroscopy screening analysis. The ranges reported for the gamma spectroscopy measurements are standard deviations derived from counting statistics.

Sample	Appearance Category	pCi/g						
		^{137}Cs	^{144}Ce	^{152}Eu	^{233}Pa	^{234}Pa	^{237}U	^{237}Np
P9GP02015G	Unknown waste	7.37 ± 0.61	$3,310 \pm 400$	—	—	—	76.4 ± 6.7	—
P9GP04015G	Unknown waste	—	—	—	—	$1,040 \pm 120$	75 ± 11	—
P9GP05015G	Unknown waste	5.45 ± 0.66	—	—	15.4 ± 3.8	205 ± 74	47.6 ± 7.1	—
P9GR04012G	Organic waste	—	7.10 ± 0.88	—	20.7 ± 1.9	—	—	—
P9GR20012G	Organic waste	3.59 ± 0.37	—	—	—	—	48.4 ± 5.4	$7,900 \pm 1,300$
P9GT01016G	Mixed soil-unknown waste	—	—	—	1.60 ± 0.23	—	6.44 ± 0.70	—
P9GT02016G	Mixed soil-unknown waste	—	—	—	16.3 ± 3.4	—	780 ± 110	—
P9GT27016G	Mixed soil-unknown waste	—	—	—	—	—	45.8 ± 6.8	—
P9GT28016G	Clean soil	—	$1,700 \pm 200$	26.6 ± 3.0	5.63 ± 0.94	—	89 ± 12	—
P9GT29016G	Clean to mostly clean soil	—	$3,320 \pm 450$	31.4 ± 4.4	4.82 ± 0.65	—	—	—
P9GT30016G	Clean to mostly clean soil	—	—	—	—	—	605 ± 89	—
P9GT31016G	Clean to mostly clean soil	—	$6,240 \pm 730$	—	—	—	—	87 ± 15
P9GT32016G	Clean to mostly clean soil	—	—	38.2 ± 4.2	5.83 ± 0.94	—	193 ± 23	—
P9GT33016G	Clean to mostly clean soil	—	—	32.3 ± 3.6	—	—	107 ± 15	—
P9GT34016G	Clean soil	—	—	—	—	—	171 ± 52	—
P9GT36016G	Clean to mostly clean soil	—	—	32.0 ± 4.0	—	—	176 ± 20	—

Appendix E

Total Actinide Analyses

Appendix E

Total Actinide Analyses

Appendix E contains the complete compilation of actinide measurements by inductively coupled plasma-mass spectroscopy (ICP-MS) (Table E-1) and calculated isotope ratios (Table E-2) for all samples. In addition, supplemental figures are provided that describe the relationship of ^{237}Np concentration to ^{241}Am concentration (Figure E-1), ^{239}Pu concentration to ^{235}U concentration (Figure E-2A), and the m/z 238/235 isotope ratio to ^{235}U concentration (Figure E-2B).

E-1. SCOPE

The total actinide analyses provide an indication of the actinides present in each sample, their concentration, and heterogeneity. The statistical mean concentration of actinides and standard deviation via analysis of three replicate subsamples of each sample. The experimental methods are presented in Section E-2. The envelope of actinides present and their concentrations are presented for all waste and soil samples in Table E-1: this is vital information that allows computation of distribution coefficients subsequent to the leaching studies described in Section 8 and Appendix F. Isotope ratios calculated for all samples are presented in Table E-2.

E-2. TOTAL ACTINIDE METHODS

Subsamples were taken from the portion of the total sample that was not used in the gamma spectroscopy screening. At least three separate aliquots of each sample were dissolved using a total dissolution procedure employing a high temperature fusion. Approximately 0.25 g of sample was first predigested in 2 mL of nitric acid and 6 drops of hydrogen peroxide and subsequently taken to dryness and “charred” on a hot plate at high setting. The sample was then placed in a furnace and ashed at 500°C. The resulting ashed soil was fused at 650–700°C with 10–15 times the sample mass of sodium peroxide in a zirconium crucible. The cake was dissolved in approximately 60 mL of deionized water and made acidic with 5 mL of concentrated nitric acid. Aliquots of the resulting solution were filtered through a 0.45 μm filter, spiked with 50 ng of indium to be used as an internal standard and diluted to 10 mL for ICP-MS.

Standards were prepared from known commercial stock solutions of thorium, depleted uranium, and lead and from neptunium and plutonium nitrate solutions in which the concentration of the analytes had been previously determined by gamma counting procedures at the RML using the standard 60 mL geometry. The resulting stock solutions for lead and neptunium were 484 ng/mL ^{239}Pu and 979 ng/mL neptunium. All ICP-MS standards were prepared from this set of stock solutions. Indium at 5 ng/mL was used as an internal standard.

The ICP-MS analyses were performed on a Thermo Electron X7 Series ICP-MS. The instrument was operated in the peak jump mode recording data for the internal standard (^{115}In), the major lead isotopes (m/z 206, 207, and 208), and the actinides (m/z 232–244). Each determination consisted of three replicates of 100 scans each and a dwell time of 10 ms. The standards for lead, ^{232}Th , ^{238}U , ^{237}Np , and ^{239}Pu were used to determine “average,” mass corrected relative response factors that were applied to the isotopic analytes determined at m/z 233–236 and 240–244. Analytes at m/z 233–236 were assumed to be uranium while plutonium was assigned to m/z 240, 242 and 244. Americium was assigned to m/z 241 and 243; however, it is believed that 10–24% of the response at m/z 241 may actually have resulted from residual ^{241}Pu . Isotope ratios were determined from the raw counts when the response for both isotopes in

the ratio were greater than three times the standard deviation of a blank which was analyzed several times during the run.

E-3. TOTAL ACTINIDE DATA

Table E-1 gives the detailed results of the total actinide analyses performed by fusion dissolution of each sample followed by analysis of the dissolution cake with an inductively coupled plasma mass spectrometer. All 36 interstitial soil samples and eight waste samples were analyzed. The data were used to calculate atom ratios, shown in Table E-2, that are useful for determining waste origins.

Table E-1. Results of total actinide analysis of sludge and interstitial soil samples. All values are reported in units of ng/g. Values reported are the averages of measurements for three subsamples, plus or minus the standard deviation. Underlined values are above or below the significance criteria, defined as the mean $\pm t_{(p=0.01)}$ standard deviation calculated for the blank INL soil (see text). Values in red are greater than, while those in blue are less than the mean of the overburden and blank soil at P=0.05.

Field Sample	Postanalysis Sample Category	Acquisition Date	Total Lead (mean)	^{232}Th	^{234}U	^{235}U	^{236}U	^{237}Np	^{238}U	^{239}Pu	^{240}Pu	^{241}Am , ^{241}Pu	^{242}Pu
Clean Soil													
P9GT13016G	Clean Soil	2/17/04	$21,200 \pm 2,000$	$11,760 \pm 690$	<0.48	28.3 ± 1.8	<1.0	<3.6	$3,760 \pm 340$	<19	0.40 ± 0.29	<0.15	<0.048
P9GT22016G	Clean Soil	2/17/04	$14,800 \pm 6,600$	$9,300 \pm 3,000$	<0.48	23.3 ± 6.2	<1.0	<3.6	$2,970 \pm 940$	<19	<0.19	<0.15	<0.048
P9GT24016G	Clean Soil	2/17/04	$22,100 \pm 1,300$	$11,900 \pm 890$	<0.48	35.0 ± 4.6	<1.0	<3.6	$3,930 \pm 410$	<19	0.95 ± 0.75	<0.15	<0.048
P9GT26016G	Clean Soil	2/17/04	$22,300 \pm 1,000$	$13,060 \pm 260$	<0.48	38.7 ± 2.8	<1.0	<3.6	$4,560 \pm 360$	<19	0.68 ± 0.40	<0.15	<0.048
Ranges for T13, T22, T24, T26	Clean Soil	—	$14,800$ –22,300	9,300–13,060	<0.48	23.3– 38.7	<1.0	<3.6	2,970– $4,560$	<19	<0.19– 0.95	<0.15	<0.048
Low-Contamination Soil													
P9GT08016G	Low-Contamination Soil	2/13/04	$23,400 \pm 2,700$	$11,710 \pm 520$	<0.48	28.0 ± 1.5	<1.0	<3.6	$3,800 \pm 220$	61 ± 30	2.4 ± 1.2	0.22 ± 0.10	<0.048
P9GT09016G	Low-Contamination Soil	2/13/04	$23,400 \pm 4,000$	$11,570 \pm 670$	<0.48	30.1 ± 2.0	<1.0	<3.6	$3,980 \pm 240$	46 ± 38	1.9 ± 1.9	0.9 ± 1.1	<0.048
P9GT10016G	Low-Contamination Soil	2/14/04	$20,900 \pm 4,600$	$12,400 \pm 1,500$	<0.48	33.7 ± 1.6	<1.0	<3.6	$3,790 \pm 420$	55 ± 21	2.22 ± 0.74	0.41 ± 0.15	<0.048
P9GT11016G	Low-Contamination Soil	2/14/04	$26,000 \pm 15,000$	$11,100 \pm 1,800$	<0.48	28.7 ± 4.8	<1.0	<3.6	$3,540 \pm 630$	74.3 ± 9.8	3.07 ± 0.64	0.63 ± 0.33	<0.048
P9GT12016G	Low-Contamination Soil	2/14/04	$23,200 \pm 3,200$	$12,710 \pm 830$	<0.48	31.9 ± 4.1	<1.0	<3.6	$3,810 \pm 340$	86 ± 40	4.2 ± 3.0	0.47 ± 0.22	0.12 ± 0.16
P9GT14016G	Low-Contamination Soil	2/17/04	$23,900 \pm 7,500$	$12,720 \pm 650$	0.79 ± 0.29	57.9 ± 9.1	<1.0	<3.6	$4,400 \pm 300$	27 ± 12	1.07 ± 0.53	<0.15	0.083 ± 0.082
P9GT15016G	Low-Contamination Soil	2/17/04	$25,300 \pm 9,600$	$12,400 \pm 1,200$	0.82 ± 0.23	77 ± 10	<1.0	<3.6	$5,070 \pm 230$	340 ± 520	3.1 ± 2.7	0.403 ± 0.090	<0.048
P9GT16016G	Low-Contamination Soil	2/17/04	$23,100 \pm 3,000$	$10,200 \pm 4,200$	0.63 ± 0.31	48 ± 15	<1.0	<3.6	$4,080 \pm 420$	21 ± 13	0.65 ± 0.48	<0.15	<0.048
P9GT18016G	Low-Contamination Soil	2/17/04	$23,900 \pm 4,700$	$13,100 \pm 3,200$	0.545 ± 0.032	67 ± 18	<1.0	<3.6	$5,000 \pm 930$	21.6 ± 3.0	0.76 ± 0.22	<0.15	0.48 ± 0.84
P9GT20016G	Low-Contamination Soil	2/17/04	$22,200 \pm 4,600$	$9,900 \pm 1,500$	0.61 ± 0.45	67 ± 25	<1.0	<3.6	$4,700 \pm 1,000$	33 ± 22	1.50 ± 0.95	<0.15	<0.048

Table E-1. (continued).

Field Sample	Postanalysis Sample Category	Acquisition Date	Total Lead (mean)	^{232}Th	^{234}U	^{235}U	^{236}U	^{237}Np	^{238}U	^{239}Pu	^{240}Pu	^{241}Am , ^{241}Pu	^{242}Pu
P9GT21016G	Low-Contamination Soil	2/17/04	<u>25,220 ± 690</u>	12,000 ± 1,100	<u>0.81 ± 0.38</u>	78 ± 19	<1.0	<3.6	<u>5,630 ± 630</u>	<u>23.0 ± 7.7</u>	<u>0.72 ± 0.50</u>	<u>0.188 ± 0.096</u>	<0.048
P9GT23016G	Low-Contamination Soil	2/17/04	17,700 ± 4,700	10,900 ± 2,000	<0.48	25.7 ± 2.6	<1.0	<3.6	3,280 ± 310	<u>23 ± 10</u>	<u>0.35 ± 0.24</u>	<u>0.16 ± 0.18</u>	<0.048
P9GT25016G	Low-Contamination Soil	2/17/04	23,000 ± 11,000	12,400 ± 3,000	<0.48	27.3 ± 6.5	<1.0	<3.6	3,630 ± 790	<u>31 ± 27</u>	<u>1.2 ± 1.1</u>	<u>0.21 ± 0.21</u>	<0.048
Ranges for T08–12, T14–T16, T18, T20–T21, T23, T25	Low-Contamination Soil	—	17,700– <u>26,000</u>	9,900–13,100	<0.48– <u>0.81</u>	25.7–78	<1.0	<3.6	3,280– <u>5,630</u>	21– <u>86</u> (T15 had a value of 340)	<u>0.35–4.2</u>	<0.15– <u>0.9</u>	<0.048– <u>0.48</u>
Mixed Soil-Waste													
P9GT01016G	Mixed Soil-Waste	2/2/04	<u>40,000 ± 37,000</u>	10,570 ± 450	<u>2.5 ± 2.8</u>	<u>190 ± 210</u>	<u>8 ± 11</u>	<u>6.6 ± 9.4</u>	<u>22,000 ± 23,000</u>	<u>260 ± 340</u>	<u>16 ± 22</u>	<u>100 ± 140</u>	<u>0.18 ± 0.25</u>
P9GT02016G	Mixed Soil-Waste	2/2/04	<u>260,000 ± 630,000</u>	10,300 ± 1,800	<u>6.9 ± 2.8</u>	<u>580 ± 230</u>	<u>24 ± 20</u>	<u>19 ± 19</u>	<u>45,000 ± 39,000</u>	<u>700 ± 660</u>	<u>43 ± 43</u>	<u>260 ± 250</u>	<u>0.24 ± 0.27</u>
P9GT03016G	Mixed Soil-Waste	2/8/04	22,600 ± 2,400	11,300 ± 1,300	<u>1.34 ± 0.45</u>	<u>141 ± 28</u>	<u>1.27 ± 0.29</u>	<3.6	<u>12,600 ± 2,100</u>	<u>3 ± 16</u>	<u>1.78 ± 0.53</u>	<u>4.2 ± 3.1</u>	<0.048
P9GT04016G	Mixed Soil-Waste	2/8/04	<u>27,900 ± 5,200</u>	10,170 ± 450	<u>1.49 ± 0.29</u>	<u>158 ± 18</u>	<1.0	<3.6	<u>10,600 ± 1,100</u>	<u>73 ± 52</u>	<u>3.5 ± 3.0</u>	<u>0.36 ± 0.11</u>	<0.048
P9GT05016G	Mixed Soil-Waste	2/8/04	<u>28,500 ± 7,300</u>	10,990 ± 920	0.67 ± 0.18	<u>72 ± 19</u>	<1.0	<3.6	<u>7,100 ± 1,400</u>	21 ± 14	<u>0.85 ± 0.27</u>	<u>4.7 ± 3.4</u>	<0.048
P9GT06016G	Mixed Soil-Waste	2/8/04	<u>40,000 ± 15,000</u>	9,800 ± 350	<u>3.9 ± 1.4</u>	<u>390 ± 120</u>	<u>2.21 ± 0.82</u>	<3.6	<u>24,400 ± 7,300</u>	<u>70 ± 32</u>	<u>2.7 ± 1.3</u>	<u>0.54 ± 0.56</u>	<0.048
P9GT07016G	Mixed Soil-Waste	2/12/04	<u>32,000 ± 3,500</u>	<u>7,400 ± 1,300</u>	<u>1.03 ± 0.17</u>	<u>111 ± 12</u>	<1.0	<3.6	<u>5,430 ± 280</u>	<u>113 ± 16</u>	<u>4.53 ± 0.54</u>	<u>0.75 ± 0.29</u>	0.057 ± 0.053
P9GT17016G	Mixed Soil-Waste	2/17/04	<u>47,000 ± 13,000</u>	<u>8,240 ± 310</u>	<u>2.03 ± 0.34</u>	<u>201 ± 41</u>	<1.0	<3.6	<u>8,500 ± 1,000</u>	<u>67 ± 36</u>	<u>3.10 ± 0.79</u>	<u>0.50 ± 0.20</u>	<0.048
P9GT19016G	Mixed Soil-Waste	2/17/04	<u>90,600 ± 8,100</u>	<u>2,200 ± 190</u>	<u>5.22 ± 0.26</u>	<u>493 ± 35</u>	<u>2.32 ± 0.36</u>	<3.6	<u>14,200 ± 1,000</u>	<u>144. ± 8.2</u>	<u>6.01 ± 0.43</u>	<u>0.54 ± 0.22</u>	<u>0.069 ± 0.035</u>
Ranges for T01–T07, T17, T19	Mixed Soil-Waste	—	22,600– <u>260,000</u>	<u>2,200</u> –11,300	<u>0.67</u> – <u>6.9</u>	<u>20</u> – <u>580</u>	<1.0– <u>24</u>	<3.6– <u>19</u>	<u>5,430</u> – <u>45,000</u>	<u>21</u> – <u>700</u>	<u>0.85</u> – <u>43</u>	<u>0.36</u> – <u>260</u>	<0.048– <u>0.24</u>
Soil Scrapped from Graphite													
P9GT27016G	Soil Scrapped from Graphite	2/17/04	<u>32,000 ± 15,000</u>	10,540 ± 800	<u>1.9 ± 1.9</u>	<u>98 ± 91</u>	<u>16 ± 19</u>	<u>17 ± 20</u>	3,520 ± 370	<u>78,000 ± 95,000</u>	<u>3,500 ± 4,000</u>	<u>300 ± 330</u>	<u>11 ± 12</u>
P9GT27016G	Soil Scrapped from Graphite	2/17/04	<u>35,500 ± 9,100</u>	10,780 ± 780	<u>0.94 ± 0.28</u>	<u>52.8 ± 6.6</u>	<u>6.09 ± 0.40</u>	<u>6.9 ± 1.4</u>	3,420 ± 370	<u>31,100 ± 1,100</u>	<u>1,469 ± 83</u>	<u>137 ± 20</u>	<u>4.88 ± 0.65</u>
Ranges for T27	Soil Scrapped from Graphite	—	<u>32,000</u> – <u>35,500</u>	10,540–10,780	<u>0.94</u> – <u>1.9</u>	<u>52.8</u> – <u>98</u>	<u>6.09</u> – <u>16</u>	<u>6.9</u> – <u>17</u>	3,420–3,520	<u>31,100</u> – <u>78,000</u>	<u>1,469</u> – <u>3,500</u>	<u>137</u> – <u>300</u>	<u>4.88</u> – <u>11</u>

Table E-1. (continued).

Field Sample	Postanalysis Sample Category	Acquisition Date	Total Lead (mean)	^{232}Th	^{234}U	^{235}U	^{236}U	^{237}Np	^{238}U	^{239}Pu	^{240}Pu	$^{241}\text{Am}, ^{241}\text{Pu}$	^{242}Pu
Soil after Rupture of Graphite Scarfings Jar													
P9GT28016G	Soil after Rupture of Graphite Scarfings Jar	2/19/04	21,800 ± 3,800	<u>13,600 ± 1,700</u>	<u>1.12 ± 0.26</u>	<u>65 ± 15</u>	<u>7.1 ± 1.5</u>	<u>7.6 ± 2.1</u>	<u>4,120 ± 740</u>	<u>35,000 ± 9,000</u>	<u>1,520 ± 380</u>	<u>130 ± 36</u>	<u>4.93 ± 0.88</u>
P9GT29016G	Soil after Rupture of Graphite Scarfings Jar	2/19/04	22,900 ± 4,500	10,900 ± 2,000	0.68 ± 0.27	<u>62 ± 18</u>	<u>6.4 ± 2.5</u>	<u>5.8 ± 1.8</u>	3,910 ± 850	<u>30,000 ± 14,000</u>	<u>1,260 ± 520</u>	<u>109 ± 46</u>	<u>4.2 ± 1.9</u>
P9GT30016G	Soil after Rupture of Graphite Scarfings Jar	2/19/04	<u>60,000 ± 110,000</u>	11,400 ± 1,500	<u>1.99 ± 0.20</u>	<u>89 ± 19</u>	<u>14.0 ± 4.5</u>	<u>14.8 ± 5.7</u>	3,930 ± 560	<u>66,000 ± 24,000</u>	<u>3,100 ± 1,000</u>	<u>280 ± 62</u>	<u>10.5 ± 2.4</u>
P9GT31016G	Soil after Rupture of Graphite Scarfings Jar	2/19/04	21,200 ± 4,900	12,180 ± 770	1.38 ± 0.34	<u>71 ± 14</u>	<u>8.4 ± 3.1</u>	<u>8.7 ± 2.8</u>	<u>4,160 ± 190</u>	<u>42,000 ± 14,000</u>	<u>1,810 ± 550</u>	<u>156 ± 47</u>	<u>6.0 ± 2.1</u>
P9GT32016G	Soil after Rupture of Graphite Scarfings Jar	2/19/04	22,900 ± 6,200	9,940 ± 350	1.55 ± 0.64	<u>85 ± 27</u>	<u>11.4 ± 5.2</u>	<u>12.9 ± 7.2</u>	3,740 ± 320	<u>60,000 ± 30,000</u>	<u>2,600 ± 1,200</u>	<u>220 ± 110</u>	<u>7.7 ± 3.0</u>
P9GT33016G	Soil after Rupture of Graphite Scarfings Jar	2/19/04	22,200 ± 1,500	10,200 ± 1,200	1.50 ± 0.95	<u>84 ± 53</u>	<u>13 ± 11</u>	<u>13 ± 11</u>	3,290 ± 300	<u>63,000 ± 59,000</u>	<u>2,800 ± 2,700</u>	<u>240 ± 220</u>	<u>9.2 ± 8.0</u>
P9GT34016G	Soil after Rupture of Graphite Scarfings Jar	2/19/04	20,700 ± 1,600	10,460 ± 490	1.37 ± 0.24	<u>83 ± 11</u>	<u>12.0 ± 1.1</u>	<u>13.30 ± 0.85</u>	3,536 ± 50	<u>61,000 ± 11,000</u>	<u>2,640 ± 440</u>	<u>231 ± 35</u>	<u>8.3 ± 1.4</u>
P9GT35016G	Soil after Rupture of Graphite Scarfings Jar	2/19/04	20,500 ± 2,600	12,120 ± 990	1.48 ± 0.49	<u>82 ± 17</u>	<u>10.1 ± 3.9</u>	<u>10.9 ± 5.0</u>	<u>4,310 ± 460</u>	<u>52,000 ± 24,000</u>	<u>2,400 ± 1,100</u>	<u>199 ± 85</u>	<u>7.3 ± 3.7</u>
P9GT36016G	Soil after Rupture of Graphite Scarfings Jar	2/19/04	<u>24,600 ± 3,100</u>	12,190 ± 850	1.27 ± 0.51	<u>90 ± 11</u>	<u>11.3 ± 3.0</u>	<u>13.8 ± 2.5</u>	<u>4,470 ± 240</u>	<u>58,000 ± 13,000</u>	<u>2,620 ± 550</u>	<u>221 ± 54</u>	<u>9.1 ± 1.7</u>
Ranges for T28–36	Soil after Rupture—of Graphite Scarfings Jar		20,500– <u>60,000</u>	9,940– <u>13,600</u>	0.68– <u>1.99</u>	<u>62–90</u>	<u>6.4–14.0</u>	<u>5.8–14.8</u>	3,290– <u>4,470</u>	<u>30,000–63,000</u>	<u>1,260–3,100</u>	<u>109–240</u>	<u>4.2–10.5</u>
Unknown Waste Type I													
P9GP01015G	Unknown Waste Type I	2/1/04	<u>2,250 ± 540</u>	<u>1,000 ± 110</u>	<0.48	<u>13.3 ± 1.1</u>	<1.0	<3.6	<u>1,500 ± 160</u>	<u>80 ± 130</u>	<u><0.19</u>	<u>0.52 ± 0.30</u>	<0.048
P9GP03015G	Unknown Waste Type I	2/1/04	<u>2,780 ± 760</u>	<u>1,020 ± 390</u>	<0.48	23 ± 18	<1.0	<3.6	2,400 ± 1,800	<u>90 ± 150</u>	<u>0.25 ± 0.35</u>	<u>4.9 ± 5.6</u>	<0.048
Ranges for P01, P03	Unknown Waste — Type I		<u>2,250–2,780</u>	<u>1,000–1,020</u>	<0.48	<u>13.3–23</u>	<1.0	<3.6	<u>1,500–2,400</u>	<u>80–90</u>	<u><0.19–0.25</u>	<u>0.52–4.9</u>	<0.048
Unknown Waste Type II													
P9GP02015G (note a)	Unknown Waste Type II	2/2/04	<u>115,000 ± 12,000</u>	<u>3,870 ± 550</u>	<u>12.3 ± 1.8</u>	<u>880 ± 110</u>	<u>56.8 ± 8.8</u>	<u>56 ± 13</u>	<u>102,000 ± 11,000</u>	<u>1,650 ± 340</u>	<u>99 ± 22</u>	<u>630 ± 120</u>	<u>0.59 ± 0.18</u>

Table E-1. (continued).

Field Sample	Postanalysis Sample Category	Acquisition Date	Total Lead (mean)	^{232}Th	^{234}U	^{235}U	^{236}U	^{237}Np	^{238}U	^{239}Pu	^{240}Pu	$^{241}\text{Am}, ^{241}\text{Pu}$	^{242}Pu
P9GP04015G	Unknown Waste Type II	2/04	$85,000 \pm 19,000$	$5,800 \pm 1,600$	6.3 ± 8.2	520 ± 250	33 ± 27	23 ± 44	$61,000 \pm 27,000$	$700 \pm 1,000$	44 ± 90	300 ± 220	0.4 ± 1.1
P9GP05015G	Unknown Waste Type II	2/04	$75,200 \pm 7,600$	$6,100 \pm 1,100$	5.8 ± 1.1	441 ± 45	29.7 ± 3.3	24.5 ± 1.8	$54,500 \pm 5,600$	930 ± 120	35 ± 31	321 ± 36	0.43 ± 0.17
Ranges for P02, P04, P05	Unknown Waste Type II	—	<u>75,200–115,000</u>	<u>3,870–6,100</u>	<u>5.8–12.3</u>	<u>441–880</u>	<u>29.7–56.8</u>	<u>23–56</u>	<u>54,500–102,000</u>	<u>700–1,650</u>	<u>35–99</u>	<u>300–630</u>	<u>0.4–0.59</u>
Organic Waste													
P9GR04012G (note b)	Organic Waste	2/1/04	$290,000 \pm 330,000$	$1,800 \pm 1,300$	35 ± 24	$2,400 \pm 1,700$	160 ± 110	137 ± 90	$220,000 \pm 110,000$	$4,900 \pm 3,700$	120 ± 110	$1,800 \pm 1,300$	2.5 ± 2.0
P9GR04012G (note c)	Organic Waste	2/1/04	$175,400 \pm 4,500$	$1,074 \pm 38$	20.6 ± 1.3	$1,442 \pm 54$	97.1 ± 5.4	84.9 ± 0.60	$159,000 \pm 22,000$	$2,790 \pm 450$	179 ± 38	$1,015 \pm 76$	1.38 ± 0.57
P9GR20012G (note d)	Organic Waste	2/2/04	$126,000 \pm 17,000$	$3,320 \pm 230$	16.9 ± 1.3	$1,410 \pm 130$	47.3 ± 6.4	36.6 ± 10.0	$142,800 \pm 6,300$	$1,230 \pm 250$	59 ± 36	454 ± 94	0.86 ± 0.41
P9GR23012G	Organic Waste	2/12/04	$122,700 \pm 7,300$	$1,780 \pm 640$	7.55 ± 0.20	772 ± 38	3.66 ± 0.40	<3.6	$19,500 \pm 1,400$	870 ± 900	25 ± 14	3.5 ± 3.2	0.17 ± 0.22
Ranges for R04, R20, R23	Organic Waste	—	<u>122,700–290,000</u>	<u>1,074–3,320</u>	<u>7.55–35</u>	<u>772–2,400</u>	<u>3.66–160</u>	<u><3.6–137</u>	<u>19,500–220,000</u>	<u>870–4,900</u>	<u>25–179</u>	<u>3.5–1,800</u>	<u>0.17–2.5</u>
Overburden Soil													
P9GW04013A	Overburden	1/27/04	$19,900 \pm 1,600$	$10,700 \pm 1,100$	<0.48	22.2 ± 1.8	<1.0	<3.6	$2,960 \pm 250$	<19	<0.19	<0.15	<0.048
P9GW09013A	Overburden	1/27/04	$20,300 \pm 2,300$	$10,120 \pm 520$	<0.48	22.2 ± 2.1	<1.0	<3.6	$3,000 \pm 330$	<19	<0.19	<0.15	<0.048
P9GW12013A	Overburden	1/27/04	$20,960 \pm 1,000$	$11,300 \pm 1,100$	<0.48	24.6 ± 1.6	<1.0	<3.6	$3,435 \pm 50$	<19	<0.19	<0.15	<0.048
P9GW13013A	Overburden	1/27/04	$19,500 \pm 3,300$	$10,270 \pm 920$	<0.48	23.2 ± 3.6	<1.0	<3.6	$3,160 \pm 400$	<19	<0.19	<0.15	<0.048
P9GW15013A	Overburden	1/29/04	$19,000 \pm 3,100$	$11,500 \pm 2,100$	<0.48	27.0 ± 8.7	<1.0	<3.6	$3,470 \pm 590$	<19	<0.19	<0.15	<0.048
P9GW21013A	Overburden	1/29/04	$21,700 \pm 9,200$	$11,000 \pm 1,300$	<0.48	29.2 ± 3.4	<1.0	<3.6	$3,380 \pm 180$	<19	<0.19	<0.15	<0.048
BLANK SOIL	INL Soil Blank	—	$20,600 \pm 1,700$	$11,860 \pm 690$	<0.48	21.4 ± 1.5	<1.0	<3.6	$2,890 \pm 200$	<19	<0.19	<0.15	<0.048
Ranges for W04, W09, W12, W13, W15, W21, Blank	Overburden, Blank	—	19,000–21,700	10,120–11,860	<0.48	21.4–29.2	<1.0	<3.6	2,890–3,435	<19	<0.19	<0.15	<0.048
a. ^{233}U was measured in P9GP02015G at 2.73 ± 0.21 ng/g.													
b. For sample P9GR04012G, all data points, including one very high replicate used in calculating average values reported. ^{233}U was measured in this sample (all data points) at 7.1 ± 5.9 ng/g.													
c. For sample P9GR04012G, all data points, <u>except</u> one very high replicate used in calculating average values reported. ^{233}U was measured in this sample (all data points <u>except</u> high replicate) at 3.73 ± 0.82 ng/g.													
d. ^{233}U was measured in P9GR20012G at 2.47 ± 0.49 ng/g.													

Table E-2. Isotope ratios in interstitial soil and waste samples. Values reported are averages of measurements for three subsamples, plus or minus the standard deviation. Ratios that are underlined are outside of the expected range found in blank INL soil. For the $^{232}\text{Th}/^{238}\text{U}$ and $^{238}\text{U}/^{235}\text{U}$ ratios, the ratios consistently fall below the blank soil average implying elevated uranium levels and ^{235}U enrichment.

Field Sample	Post-Analysis Category	Atom Ratios						
		232/238	238/235	238/239	239/238	239/240	238/236	241/237
Clean Soil								
P9GT13016G	Clean Soil	3.039 ± 0.091	134.5 ± 3.5	<u>600 ± 320</u>	<u>0.0020 ± 0.0010</u>	<u>16.7 ± 1.3</u>	<u>$75,000$</u>	—
P9GT22016G	Clean Soil	3.01 ± 0.29	128.1 ± 6.8	—	—	—	<u>$60,000$</u>	—
P9GT24016G	Clean Soil	2.94 ± 0.10	<u>114.3 ± 3.7</u>	—	—	—	<u>$66,600 \pm 5,900$</u>	—
P9GT26016G	Clean Soil	2.78 ± 0.24	119.6 ± 7.4	<u>350</u>	<u>0.0029</u>	<u>18</u>	<u>$90,000 \pm 110,000$</u>	—
Ranges for T13, T22, T24, T26	Clean Soil	2.78–3.04	<u>114.3</u> – <u>134.5</u>	<u>350</u> – <u>600</u>	<u>0.0020</u> – <u>0.0029</u>	<u>16.7</u> – <u>18</u>	<u>$66,600$</u> – <u>$90,000$</u>	—
Low-Contamination Soil								
P9GT08016G	Low-Contamination Soil	2.99 ± 0.15	137.9 ± 1.6	<u>98 ± 45</u>	<u>0.0117 ± 0.0051</u>	<u>19.37 ± 0.48</u>	<u>$96,600 \pm 4,900$</u>	—
P9GT09016G	Low-Contamination Soil	2.82 ± 0.13	134.3 ± 1.6	<u>180 ± 120</u>	<u>0.0083 ± 0.0065</u>	<u>22.7 ± 8.1</u>	<u>$63,000 \pm 37,000$</u>	—
P9GT10016G	Low-Contamination Soil	2.86 ± 0.22	119.6 ± 7.6	<u>107 ± 43</u>	<u>0.0102 ± 0.0037</u>	<u>18.79 ± 0.96</u>	—	—
P9GT11016G	Low-Contamination Soil	3.070 ± 0.068	124.8 ± 3.8	<u>70 ± 16</u>	<u>0.0147 ± 0.0030</u>	<u>17.4 ± 1.9</u>	<u>$60,100 \pm 9,400$</u>	—
P9GT12016G	Low-Contamination Soil	2.99 ± 0.23	126.4 ± 8.2	<u>63 ± 22</u>	<u>0.0180 ± 0.0082</u>	<u>16.2 ± 2.6</u>	<u>$20,000$</u>	—
P9GT14016G	Low-Contamination Soil	2.43 ± 0.27	<u>83.2 ± 8.3</u>	<u>280 ± 170</u>	<u>0.0044 ± 0.0021</u>	<u>21.5 ± 6.5</u>	<u>$42,000$</u>	—
P9GT15016G	Low-Contamination Soil	<u>2.10 ± 0.12</u>	<u>70.7 ± 4.6</u>	<u>117 ± 96</u>	<u>0.050 ± 0.077</u>	<u>54 ± 52</u>	<u>$6,700$</u>	—
P9GT16016G	Low-Contamination Soil	<u>2.02 ± 0.62</u>	<u>95 ± 17</u>	<u>440 ± 370</u>	<u>0.0034 ± 0.0020</u>	<u>28 ± 13</u>	<u>$110,000$</u>	—
P9GT18016G	Low-Contamination Soil	2.32 ± 0.11	<u>79.8 ± 4.8</u>	<u>344 ± 80</u>	<u>0.00303 ± 0.00080</u>	<u>17.9 ± 2.2</u>	<u>$14,300 \pm 3,100$</u>	—
P9GT20016G	Low-Contamination Soil	<u>2.07 ± 0.25</u>	<u>74 ± 12</u>	—	—	—	<u>$18,600 \pm 6,600$</u>	—
P9GT21016G	Low-Contamination Soil	<u>2.07 ± 0.25</u>	<u>75 ± 11</u>	<u>394 ± 99</u>	<u>0.00265 ± 0.00068</u>	—	<u>$19,000 \pm 10,000$</u>	—
P9GT23016G	Low-Contamination Soil	3.18 ± 0.32	129.4 ± 3.6	<u>232 ± 97</u>	<u>0.0049 ± 0.0020</u>	—	<u>$29,200 \pm 1,200$</u>	—
P9GT25016G	Low-Contamination Soil	3.29 ± 0.16	135.6 ± 3.5	<u>170 ± 110</u>	<u>0.0076 ± 0.0049</u>	<u>20</u>	<u>$49,000 \pm 13,000$</u>	—
Ranges for T08-12, T14-16, T18,	Low-Contamination Soil	<u>2.02</u> –3.29	<u>70.7</u> –137.9	<u>63</u> –440	<u>0.00265</u> – <u>0.0180</u>	<u>16.2</u> –54	<u>$6,700$</u> – <u>$110,000$</u>	—
—								
42–470								

Table E-2. (continued).

Field Sample	Post-Analysis Category	Atom Ratios								
		232/238	238/235	238/239	239/238	239/240	238/236	241/237	239/241	
T20-21, T23, T25										
Mixed Soil-Waste										
P9GT01016G	Mixed Soil-Waste	<u>0.97 ± 0.73</u>	<u>117.4 ± 3.8</u>	<u>200 ± 120</u>	<u>0.0064 ± 0.0032</u>	<u>17.2 ± 8.7</u>	<u>4,300 ± 1,800</u>	<u>18</u>	<u>2.0</u>	
P9GT02016G	Mixed Soil-Waste	<u>0.36 ± 0.24</u>	<u>77 ± 41</u>	<u>97 ± 16</u>	<u>0.0105 ± 0.0016</u>	<u>12.40 ± 0.72</u>	<u>1,830 ± 260</u>	<u>16.84 ± 0.68</u>	<u>2.0</u>	
P9GT03016G	Mixed Soil-Waste	<u>0.88 ± 0.20</u>	<u>92 ± 14</u>	<u>520 ± 180</u>	<u>0.00209 ± 0.00068</u>	<u>15.7 ± 3.3</u>	<u>10,180 ± 750</u>	—	<u>8.3</u>	
P9GT04016G	Mixed Soil-Waste	<u>0.93 ± 0.11</u>	<u>68.1 ± 1.0</u>	<u>260 ± 140</u>	<u>0.0051 ± 0.0037</u>	<u>17.1 ± 2.5</u>	<u>14,400 ± 3,900</u>	—	<u>160</u>	
P9GT05016G	Mixed Soil-Waste	<u>1.52 ± 0.18</u>	<u>102 ± 12</u>	<u>480 ± 210</u>	<u>0.00232 ± 0.00097</u>	<u>20.8 ± 4.3</u>	<u>9,900 ± 3,900</u>	—	—	
P9GT06016G	Mixed Soil-Waste	<u>0.42 ± 0.15</u>	<u>64.0 ± 1.5</u>	<u>510 ± 110</u>	<u>0.00201 ± 0.00041</u>	<u>20.1 ± 3.9</u>	<u>11,500 ± 1,100</u>	—	<u>89</u>	
P9GT07016G	Mixed Soil-Waste	<u>1.31 ± 0.19</u>	<u>49.9 ± 5.3</u>	<u>66.3 ± 8.8</u>	<u>0.0153 ± 0.0021</u>	<u>18.9 ± 1.2</u>	<u>10,800 ± 1,100</u>	—	<u>120</u>	
P9GT17016G	Mixed Soil-Waste	<u>0.940 ± 0.081</u>	<u>44.2 ± 9.7</u>	<u>199 ± 73</u>	<u>0.0057 ± 0.0026</u>	<u>15.9 ± 3.6</u>	<u>12,300 ± 3,700</u>	—	<u>130</u>	
P9GT19016G	Mixed Soil-Waste	<u>0.1500 ± 0.0046</u>	<u>29.3 ± 1.1</u>	<u>136 ± 18</u>	<u>0.0075 ± 0.0011</u>	<u>18.22 ± 0.72</u>	<u>6,400 ± 1,500</u>	—	<u>190</u>	
Ranges for T01-07, T17, T19	Mixed Soil-Waste	<u>0.150–1.52</u>	<u>29.3–117.4</u>	<u>97–520</u>	<u>0.00201–0.0153</u>	<u>12.40–20.8</u>	<u>1,830–14,400</u>	<u>16.84–18</u>	<u>2.0–190</u>	
Soil Scraped from Graphite										
P9GT27016G	Soil Scraped from Graphite	<u>2.93 ± 0.35</u>	<u>53 ± 24</u>	<u>0.120 ± 0.065</u>	<u>16 ± 18</u>	<u>16.26 ± 0.95</u>	<u>450 ± 240</u>	<u>23.9 ± 3.2</u>	<u>200</u>	
P9GT27016G	Soil Scraped from Graphite	<u>3.09 ± 0.19</u>	<u>65.4 ± 2.8</u>	<u>0.152 ± 0.012</u>	<u>6.60 ± 0.53</u>	<u>15.79 ± 0.13</u>	<u>569 ± 35</u>	<u>25.2 ± 2.2</u>	<u>170</u>	
Ranges for T27	Soil Scraped from Graphite	<u>2.93–3.09</u>	<u>53–65.4</u>	<u>0.120–0.152</u>	<u>6.60–16</u>	<u>15.79–16.26</u>	<u>450–569</u>	<u>23.9–25.2</u>	<u>170–200</u>	
Soil after Rupture of Graphite Scarfings Jar										
P9GT28016G	Soil after Rupture of Graphite Scarfings Jar	<u>3.24 ± 0.18</u>	<u>65.0 ± 7.4</u>	<u>0.165 ± 0.030</u>	<u>6.2 ± 1.1</u>	<u>17.30 ± 0.21</u>	<u>596 ± 93</u>	<u>21.1 ± 1.7</u>	<u>200</u>	
P9GT29016G	Soil after Rupture of Graphite Scarfings Jar	<u>2.70 ± 0.14</u>	<u>66 ± 11</u>	<u>0.202 ± 0.095</u>	<u>5.6 ± 2.1</u>	<u>17.6 ± 1.1</u>	<u>± 240</u>	<u>24.6 ± 8.9</u>	<u>200</u>	
P9GT30016G	Soil after Rupture of Graphite Scarfings Jar	<u>2.80 ± 0.17</u>	<u>45.6 ± 8.0</u>	<u>0.086 ± 0.024</u>	<u>12.4 ± 3.9</u>	<u>16.27 ± 0.60</u>	<u>300 ± 78</u>	<u>23.4 ± 3.6</u>	<u>180</u>	

Table E-2. (continued).

Field Sample	Post-Analysis Category	Atom Ratios							
		232/238	238/235	238/239	239/238	239/240	238/236	241/237	239/241
P9GT31016G	Soil after Rupture of Graphite Scarfings Jar	2.826 ± 0.058	<u>61 ± 12</u>	<u>0.144 ± 0.050</u>	<u>7.5 ± 2.5</u>	<u>17.85 ± 0.53</u>	<u>550 ± 200</u>	<u>21.5 ± 1.0</u>	—
P9GT32016G	Soil after Rupture of Graphite Scarfings Jar	2.58 ± 0.14	<u>48 ± 19</u>	<u>0.106 ± 0.065</u>	<u>12.1 ± 7.1</u>	<u>17.39 ± 0.41</u>	<u>400 ± 220</u>	<u>21.7 ± 1.7</u>	<u>200</u>
P9GT33016G	Soil after Rupture of Graphite Scarfings Jar	3.01 ± 0.12	<u>49 ± 23</u>	<u>0.117 ± 0.074</u>	<u>14 ± 13</u>	<u>16.59 ± 0.17</u>	<u>390 ± 230</u>	<u>21.0 ± 1.6</u>	<u>190</u>
P9GT34016G	Soil after Rupture of Graphite Scarfings Jar	2.86 ± 0.17	<u>43.8 ± 5.6</u>	<u>0.082 ± 0.016</u>	<u>12.5 ± 2.2</u>	<u>17.13 ± 0.36</u>	<u>300 ± 26</u>	<u>21.2 ± 2.4</u>	<u>200</u>
P9GT35016G	Soil after Rupture of Graphite Scarfings Jar	2.73 ± 0.23	<u>56 ± 17</u>	<u>0.133 ± 0.071</u>	<u>9.4 ± 5.4</u>	<u>17.03 ± 0.15</u>	<u>500 ± 260</u>	<u>22.4 ± 1.8</u>	<u>200</u>
P9GT36016G	Soil after Rupture of Graphite Scarfings Jar	2.63 ± 0.14	<u>50.6 ± 4.2</u>	<u>0.107 ± 0.022</u>	<u>9.6 ± 1.8</u>	<u>17.00 ± 0.39</u>	<u>420 ± 110</u>	<u>19.4 ± 2.9</u>	<u>200</u>
Ranges for T28-36	Soil after Rupture of Graphite Scarfings Jar	2.58–3.24	<u>43.8–66</u>	<u>0.082–0.165</u>	<u>5.6–14</u>	<u>16.27–17.85</u>	<u>300–670</u>	<u>19.4–24.6</u>	<u>180–210</u>
Unknown Waste Type I									
P9GP01015G	Unknown Waste Type I	<u>0.636 ± 0.072</u>	<u>115.3 ± 2.4</u>	<u>8.4</u>	<u>0.12</u>	<u>17</u>	<u>4,900</u>	—	<u>120</u>
P9GP03015G	Unknown Waste Type I	<u>0.48 ± 0.18</u>	<u>114.0 ± 5.6</u>	<u>230 ± 300</u>	<u>0.023 ± 0.030</u>	<u>24 ± 13</u>	<u>3,800 ± 1,300</u>	—	<u>17</u>
Ranges for P01, P03	Unknown Waste Type I	<u>0.48–0.636</u>	<u>114.0–115.3</u>	<u>8.4–230</u>	<u>0.023–0.12</u>	<u>17–24</u>	<u>3,800–4,900</u>	—	<u>17–120</u>
Unknown Waste Type II									
P9GP02015G	Unknown Waste Type II	<u>0.0380 ± 0.0079</u>	<u>116.9 ± 5.0</u>	<u>86.4 ± 8.0</u>	<u>0.0117 ± 0.0012</u>	<u>12.29 ± 0.16</u>	<u>1,819 ± 86</u>	<u>14.33 ± 0.85</u>	<u>1.9</u>
P9GP04015G	Unknown Waste Type II	<u>0.095 ± 0.025</u>	<u>116.4 ± 4.6</u>	<u>111 ± 83</u>	<u>0.0090 ± 0.0067</u>	<u>12.6 ± 4.1</u>	<u>1,860 ± 370</u>	<u>16</u>	<u>1.9</u>
P9GP05015G	Unknown Waste Type II	<u>0.1,061 ± 0.0071</u>	<u>126.1 ± 1.2</u>	<u>81 ± 15</u>	<u>0.0126 ± 0.0021</u>	<u>12.32 ± 0.66</u>	<u>1,881 ± 12</u>	<u>16.1 ± 1.7</u>	<u>2.3</u>
Ranges for P02, P04, P05	Unknown Waste Type II	<u>0.0380–0.1,061</u>	<u>116.4–126.1</u>	<u>81–111</u>	<u>0.0090–0.0126</u>	<u>12.29–12.6</u>	<u>1,819–1,860</u>	<u>14.33–16.1</u>	<u>1.9–2.3</u>
Organic Waste									
P9GR04012G	Organic Waste	<u>0.0075 ± 0.0014</u>	<u>100 ± 19</u>	<u>69 ± 17</u>	<u>0.0155 ± 0.0043</u>	<u>11.92 ± 0.65</u>	<u>1,490 ± 270</u>	<u>15.28 ± 0.88</u>	<u>2.2</u>
P9GR04012G	Organic Waste	<u>0.0069 ± 0.0015</u>	<u>110 ± 13</u>	<u>78.66 ± 0.53</u>	<u>0.01302 ± 0.00035</u>	<u>11.62 ± 0.54</u>	<u>1,640 ± 160</u>	<u>14.88 ± 0.74</u>	<u>2.0</u>

Table E-2. (continued).

Field Sample	Post-Analysis Category	Atom Ratios							
		232/238	238/235	238/239	239/238	239/240	238/236	241/237	239/241
P9GR20012G	Organic Waste	<u>0.0224 ± 0.0024</u>	<u>103.1 ± 6.2</u>	<u>163 ± 30</u>	<u>0.0063 ± 0.0013</u>	<u>12.634 ± 0.032</u>	<u>3.120 ± 440</u>	<u>15.5 ± 1.1</u>	<u>2.0</u>
P9GR23012G	Organic Waste	<u>0.087 ± 0.027</u>	<u>25.55 ± 0.78</u>	<u>59 ± 43</u>	<u>0.033 ± 0.035</u>	<u>16.0 ± 1.8</u>	<u>5.470 ± 400</u>	—	<u>180</u>
Ranges for R04, R20, R23	Organic Waste	<u>0.0069–0.087</u>	<u>25.55–110</u>	<u>59–163</u>	<u>0.0063–0.033</u>	<u>11.62–16.0</u>	<u>1,490–5,470</u>	<u>14.88–15.5</u>	<u>2.0–180</u>
Overburden Soil									
P9GW04013A	Overburden Soil	3.51 ± 0.18	136.0 ± 4.1	—	—	—	—	—	
P9GW09013A	Overburden Soil	3.28 ± 0.26	137.76 ± 0.80	—	—	—	—	—	
P9GW12013A	Overburden Soil	3.20 ± 0.23	141.7 ± 7.4	—	—	—	—	—	
P9GW13013A	Overburden Soil	3.16 ± 0.27	139 ± 11	—	—	—	—	—	
P9GW15013A	Overburden Soil	3.25 ± 0.33	149 ± 24	—	—	—	—	—	
P9GW21013A	Overburden Soil	3.17 ± 0.29	131 ± 15	—	—	—	—	—	
BLANK Soil	INL Blank Soil	4.006 ± 0.076	137.2 ± 8.7	—	—	—	—	—	
Ranges for W04, W09, W12, W13, W15, W21, Blank	INL Blank Soil	<u>3.16–4.006</u>	<u>131–141.7</u>	—	—	—	—	—	

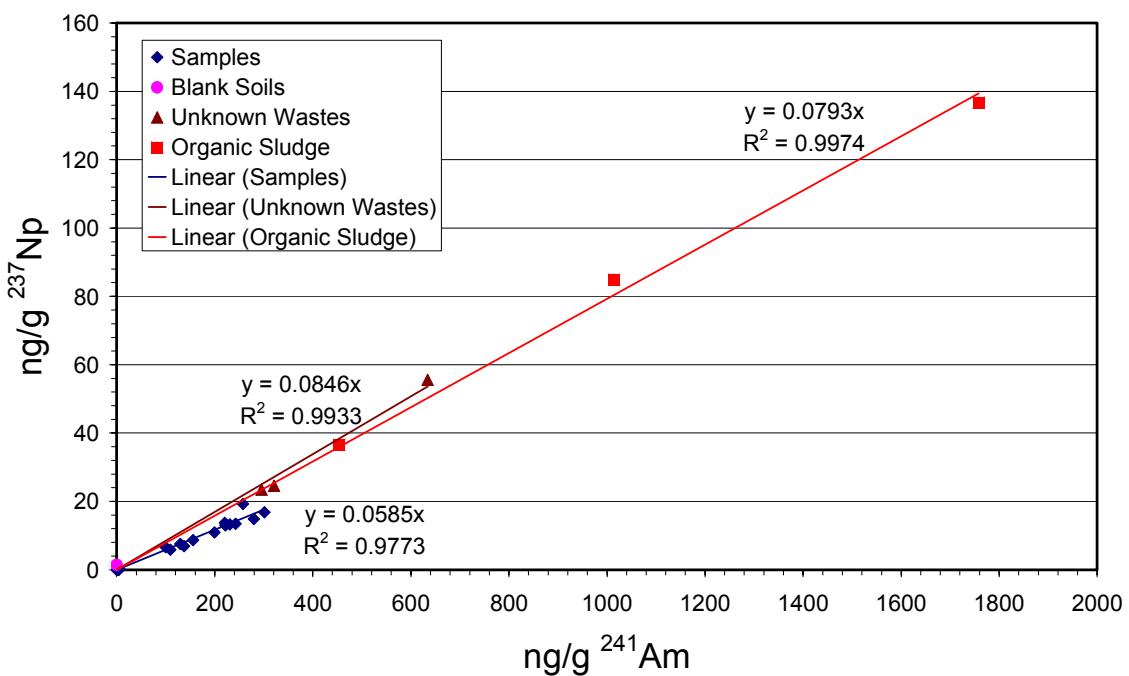


Figure E-1. Plot of ^{237}Np vs ^{241}Am for all soil and sludge samples and wastes. Lines represent the linear correlation between ^{237}Np and ^{241}Am for each sample type.

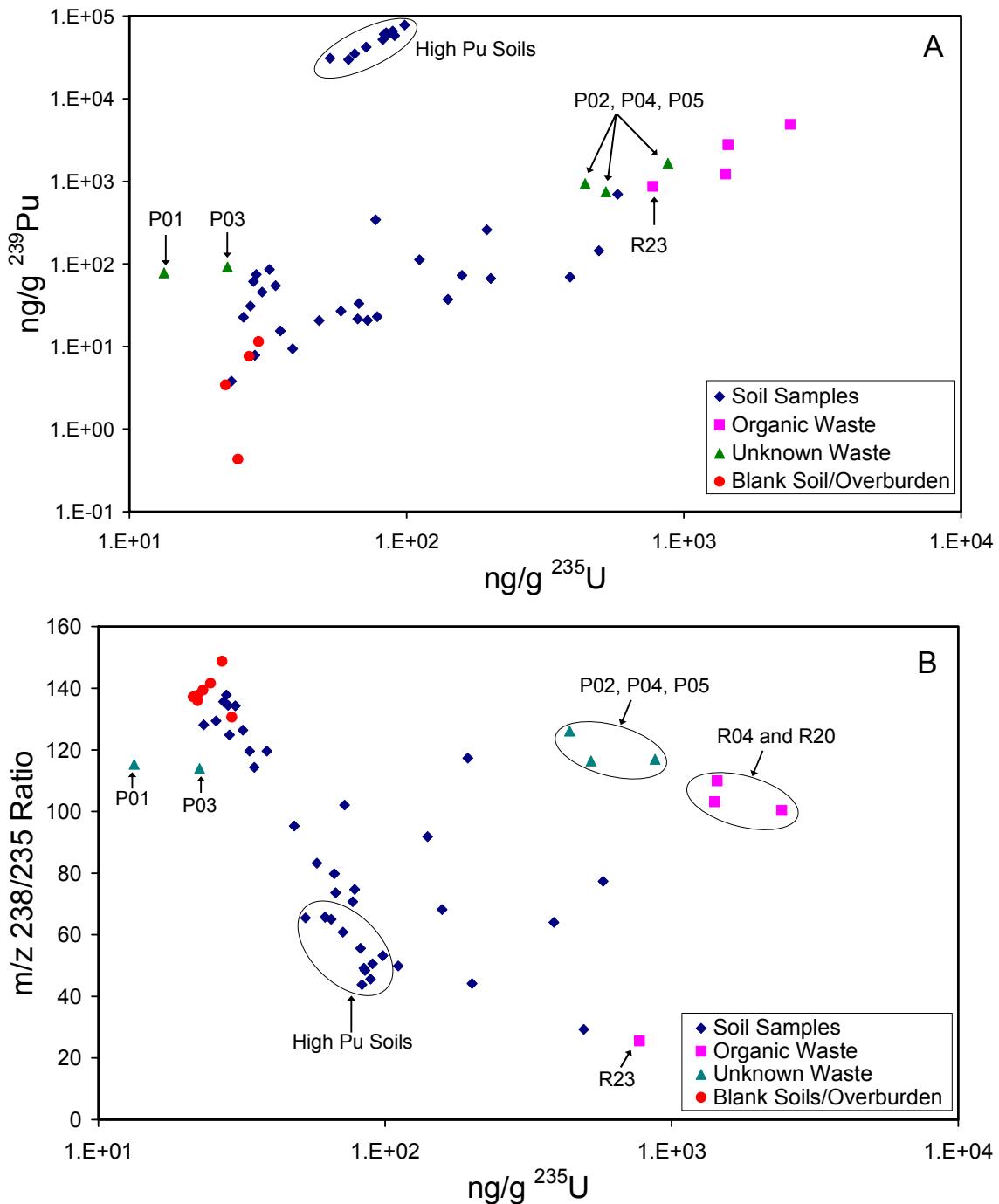


Figure E-2. Plots of (A) $[^{239}\text{Pu}]$ versus $[^{235}\text{U}]$, and (B) $^{238}\text{U}/^{235}\text{U}$ ratio versus $[^{235}\text{U}]$. High-plutonium soil sample include the soil scraped from the graphite and the soil samples after rupture of graphite scarfings jar, and have a $^{238}\text{U}/^{235}\text{U}$ range of 40–70 (circled diamond points in 11B). $^{238}\text{U}/^{235}\text{U}$ ratios for the waste samples are much less enriched (except for R23), ranging from 90 to 130 (circled triangle and square points in 11B).

Appendix F

Leaching Studies

Appendix F

Leaching Studies

Aqueous partitioning of radionuclides can be significantly enhanced by minor changes in the soil chemical environment. Alterations in soil pH and ionic strength can be brought about by many different factors, which can be either anthropic or natural (e.g., cyclic wetting and drying, intrusion of chemical agents, or microbial action). These alterations can in turn cause changes in the aqueous partitioning, which is quantitatively described using an operational distribution coefficient that is defined in the present context as a K_d (see discussion in section 8). The K_d values can be used to evaluate release and readsorption of actinides from soil and waste. Solid-solution partitioning data collected for all samples are presented in Tables F-1 and F-2.

F-1. EXPERIMENTAL PROCEDURE FOR LEACHING ANALYSES

Three different 1-g aliquots of each sample were weighed into separate 15-mL graduated centrifuge tubes. Ten mL of either deionized water or a 100 mM NaCl solution was added to each with a calibrated 10-mL pipette. The samples were stirred on a vortex mixer to suspend the solids and the samples were then rotated on a rotary mixer for about 18 hours. Samples were then centrifuged at 1,200 rpm for 20 minutes to clarify the solution. The pH for each sample was recorded at this point and 1 mL of the aqueous phase was removed for analysis. One mL of either deionized water or the 100 mM NaCl solution was added back to the tube to maintain the volume at 10 mL. The solutions were made either more acidic or basic by adding an aliquot of either concentrated nitric acid or 10% sodium hydroxide to each sample. The samples were allowed to off-gas for about 15 minutes prior to capping and vortex mixing to resuspend solids into the aqueous phase. Each sample was then vented again for about 10 minutes to ensure there would be no gaseous buildup. Each sample was then placed on the rotary mixer for about 18 hours, then centrifuged, the pH measured, and an aliquot taken for analysis. The entire process was repeated until the pH was <3 or >9.5 .

Some samples needed smaller or larger amounts of nitric acid or 10% NaOH to achieve the desired pH change due to the inherent buffering capacity of the solid materials. The organic sludge samples (R04, R20 and R23) were done in a similar fashion, using different amounts of acid and/or base because these samples had initial pH values as high as 11.7. There were sometimes significant differences in the pH readings in the replicate samples due to the inherent heterogeneity of the sample. A sample of the leachate was taken if a significant change (e.g. at least 0.8 pH units) in pH was achieved.

Leachates acquired for ICP-MS analysis were diluted and prepared by addition of 100 uL of 500 ppb indium internal standard spiking solution followed by addition of 100 uL of Optima concentrated nitric acid (1% final minimum acid concentration) and subsequent dilution to 10 mL. The resulting 10-mL solutions were then filtered through a 0.45 μm PTFE filter into new, clean graduated 15-mL tubes.

The complete results of the leaching analyses are summarized in Tables F-1 and F-2. The relatively large standard deviations reflect the sample heterogeneity, analytical uncertainty (control limits were 10–15%), and, for some samples, the sometimes very low concentrations of the analytes detected in the aqueous phase. The complete sets of K_d for all pH values are

represented graphically in Figures F-1 thru F-8. The uncertainty in defining a K_d is clearly represented in these figures as the range can be nearly and order of magnitude in most cases.

F-2. SUMMARY OF K_d VALUES

This section summarizes the K_d values for the leaching analyses in tabular and graphic format. Table F-1 shows the initial K_d value determined from the equilibration of 10 mL deionized water with about 1 g of sample for the elements ^{235}U , ^{237}Np , ^{238}U , ^{239}Pu , ^{240}Pu , ^{241}Am , thorium, and lead. Table F-2 summarizes the K_d value determined from the equilibration of 10 mL 100 mM NaCl solution with about 1 g of sample. Figures F-1 through F-8 graphically represent the K_d values for the elements (listed above) determined for the various interstitial soil samples, organic sludge samples, overburden soil samples, and a blank and plutonium-spiked blank soil sample with deionized water (top) and a 100 mM NaCl solution (bottom) as the leachate.

Table F-1. Initial K_d value determined from the equilibration of 10-mL deionized water with about 1 g of sample. Values from the total actinide analyses in Appendix E were used as the total actinide concentration. Values are ± 1 standard deviation and do not include errors that could be propagated from Appendix E. Isotope ratios were determined directly from the intensity values at the associated m/z values after subtraction of the mean blank values. In all cases, $n=6$, however, only K_d values determined when the analyte concentration was above the detection limit are included in the averages. Note the sample number has decreased as a result of the sample downselection process. If the K_d value does not include a \pm value, it is because only one measurement was available.

Sample	pH	K_d (mL/g)								Atom Ratio	
		Lead	^{232}Th	^{235}U	^{237}Np	^{238}U	^{239}Pu	^{240}Pu	^{241}Am	238/235	239/240
Clean Soil											
Range for Clean Soil		4,580–16,700	56,000–160,000	675–2,860	—	2,110–4,260	—	—	—	—	—
P9GT13	8.76 \pm 0.05	6,800 \pm 1,600	56,000 \pm 19,000	2,860 \pm 590	—	4,260 \pm 380	—	—	—	103 \pm 13	—
P9GT22	8.82 \pm 0.04	4,580 \pm 650	87,000 \pm 32,000	1,760 \pm 450	—	3,040 \pm 260	—	—	—	71 \pm 12	—
P9GT24	8.78 \pm 0.04	6,600 \pm 1,500	91,000 \pm 72,000	675 \pm 93	—	2,110 \pm 130	—	—	—	38.5 \pm 3.2	—
P9GT26	8.52 \pm 0.17	16,700 \pm 8,200	160,000 \pm 170,000	1,077 \pm 81	—	3,240 \pm 410	—	—	—	42.3 \pm 3.9	—
Low-Concentration Soil											
Range for Low-Concentration Soil		4,190–17,100	13,000–364,000	660–5,300	—	1,090–8,900	970–43,000	2,177–7,200	910–1,300	—	—
P9GT08	8.53 \pm 0.12	8,700 \pm 1,300	364,000 \pm 79,000	5,300 \pm 1,100	—	8,900 \pm 1,400	7,200	—	—	87.2 \pm 8.0	—
P9GT10	8.61 \pm 0.07	4,500 \pm 1,300	116,500 \pm 9,300	732 \pm 96	—	1,580 \pm 130	—	7,200 \pm 1,900	1,300	60.7 \pm 4.0	—
P9GT11	8.74 \pm 0.04	5,390 \pm 770	114,000 \pm 32,000	1,060 \pm 150	—	2,170 \pm 190	7,400 \pm 6,300	6,200 \pm 4,200	910 \pm 590	64.9 \pm 3.7	15
P9GT12	8.54 \pm 0.09	4,190 \pm 220	156,000 \pm 37,000	1,410 \pm 150	—	2,640 \pm 260	8,700 \pm 7,400	6,600 \pm 3,600	—	64.6 \pm 4.5	10.31 \pm 0.90
P9GT14	9.06 \pm 0.05	14,000 \pm 5,400	—	960 \pm 180	—	2,390 \pm 140	970 \pm 520	2,177 \pm 28	—	35.6 \pm 5.3	69 \pm 33
P9GT15	9.16 \pm 0.08	14,100 \pm 8,000	13,000	660 \pm 110	—	1,090 \pm 130	43,000	—	—	46.7 \pm 2.2	—
P9GT16	8.99 \pm 0.09	14,800 \pm 4,100	—	848 \pm 92	—	2,720 \pm 410	2,420 \pm 350	—	—	31.3 \pm 3.8	—
P9GT18	8.89 \pm 0.20	17,100 \pm 4,500	241,000 \pm 60,000	850 \pm 130	—	1,580 \pm 240	—	4,100	—	41.0 \pm 2.2	—
Mixed-Soil Waste											
Range for Mixed Soil Wastes		7,500–43,000	34,000–450,000	180–11,900	—	2,250–18,000	2,400–15,000	6,900–10,500	2,020–6,400	—	—
P9GT03	8.93 \pm 0.13	43,000 \pm 9,700	326,000 \pm 69,000	950 \pm 510	—	2,250 \pm 750	2,400 \pm 1,300	—	6,400 \pm 3,900	40.9 \pm 8.5	—
P9GT05	9.45 \pm 0.10	28,800 \pm 4,400	450,000	11,900 \pm 3,100	—	15,300 \pm 4,500	—	—	—	83 \pm 12	—
P9GT07	7.97 \pm 0.19	11,200 \pm 2,700	225,000 \pm 90,000	180 \pm 130	—	410 \pm 280	3,200 \pm 1,100	—	—	21.5 \pm 1.9	—
P9GT17	8.60 \pm 0.13	7,500 \pm 4,700	180,000 \pm 110,000	1,720 \pm 880	—	3,500 \pm 2,200	5,900 \pm 5,000	10,500 \pm 7,200	2,020 \pm 300	32 \pm 31	40 \pm 31
P9GT19	8.76 \pm 0.01	9,300 \pm 5,300	34,000 \pm 14,000	8,300 \pm 4,700	—	18,000 \pm 11,000	15,000	6,900 \pm 2,900	—	14.7 \pm 3.3	9.9
Soil Scrapped From Graphite											
P9GT27	8.23 \pm 0.03	32,000 \pm 26,000	430,000 \pm 200,000	1,050 \pm 590	1,700 \pm 1,800	1,950 \pm 760	730,000 \pm 80,000	430,000 \pm 140,000	57,000 \pm 35,000	35 \pm 12	20.0 \pm 3.7

Table F-1. (continued).

Sample	pH	K_d (mL/g)								Atom Ratio	
		Lead	^{232}Th	^{235}U	^{237}Np	^{238}U	^{239}Pu	^{240}Pu	^{241}Am	238/235	239/240
Soil After Rupture of Graphite Scarfings Jar											
Range for Soil After Rupture of Graphite Scarfings Jar		9,100–12,400	98,000–240,000	4,080–8,300	—	4,730–8,830	2,640,000–1,450,000	760,000–2,570,000	22,600–88,000	—	—
P9GT28	8.69±0.02	9,100±3,400	137,000±28,000	4,080±950	—	4,730±610	1,450,000±350,000	760,000	22,600±7,100	53.9 ± 7.1	13
P9GT31	8.77±0.09	12,400±8,500	102,000±14,000	4,520±380	—	5,500±590	1,510,000±840,000	1,474,000±14,000	29,000±10,000	48.3 ± 5.9	39 ± 12
P9GT32	8.75±0.04	10,070±560	98,000±21,000	7,500±1,300	—	7,400±640	2,640,000±650,000	2,030,000±660,000	57,000±23,000	44.3 ± 4.8	18.7 ± 7.9
P9GT33	8.65±0.04	11,600±1,600	182,000±20,000	7,000±1,000	—	5,390±220	4,030,000±790,000	2,350,000±780,000	85,000±16,000	50.8 ± 7.7	14.5 ± 2.8
P9GT34	8.75±0.07	9,700±1,400	107,000±18,000	6,800±1,800	—	6,510±500	2,080,000±580,000	1,420,000±360,000	56,000±36,000	44 ± 11	16.55 ± 0.47
P9GT35	8.68±0.12	9,110±650	119,000±31,000	6,220±650	—	6,850±350	2,110,000±570,000	1,520,000±530,000	34,000±7,700	47.8 ± 5.0	17.0 ± 4.8
P9GT36	8.58±0.08	11,200±2,400	240,000±150,000	8,300±1,900	—	8,830±560	4,000,000±1,000,000	2,570,000±550,000	88,000±51,000	46.9 ± 9.8	15.2 ± 6.6
Organic Waste											
Range for Organic Wastes		9,900–66,000	35,000–74,000	8,500–35,600	—	21,800–39,000	—	—	59,000–68,000	—	—
P9GR04	11.78±0.04	21,300±3,400	35,000	24,700±4,600	—	24,200±2,600	—	67,000	68,000	118 ± 13	—
P9GR20	9.76±0.12	66,000±26,000	74,000	35,600±7,300	—	39,000±12,000	—	—	59,000±21,000	112 ± 20	—
P9GR23	8.90±0.05	9,900±6,000	70,000±26,000	8,500±3,800	—	21,800±9,000	—	—	—	10.1 ± 1.1	—
Overburden Soil											
Range for Overburden Soil Samples		45,400–22,000	42,000–430,000	1,500–400	—	5,314–10,500	—	—	—	—	—
P9GW09	9.14±0.05	4,540±720	42,000±13,000	3,930±360	—	5,930±360	—	—	—	93 ± 12	—
P9GW13	9.13±0.01	8,600±3,200	30,000±13,000	3,900±1,200	—	6,900±200	—	—	—	81 ± 25	—
P9GW15	9.12±0.03	22,000±20,000	100,000±110,000	4,900±2,600	—	8,100±4,200	—	—	—	89 ± 12	—
Blank Soil	8.45±0.07	14,960±720	58,000±6,900	—	—	5,314±81	—	—	—	—	—
Pu Spiked Soil	7.88±0.06	4,960±430	430,000±160,000	1,500±1,200	—	10,500±7,300	12,400±1,800	11,300±3,000	—	88 ± 16	105.2 ± 3.5

Table F-2. Initial K_d value determined from the equilibration of 10 mL 100 mM NaCl solution with about 1 g of sample. Values from the total actinide analyses in Appendix E were used as the total actinide concentration. Values are ± 1 standard deviation and do not include errors that could be propagated from Appendix E. Isotope ratios were determined directly from the intensity values at the associated m/z values after subtraction of the mean blank values. In all cases, $n=6$, however, only K_d values determined when the analyte concentration was above the detection limit are included in the averages. Note sample number has decreased as a result of the sample downselection process. If the K_d value does not include a \pm value, it is because only one measurement was available.

Sample	pH	K_d (mL/g)								Atom Ratio	
		Pb	^{232}Th	^{235}U	^{237}Np	^{238}U	^{239}Pu	^{240}Pu	^{241}Am	$^{238}/^{235}$	$^{239}/^{240}$
Clean Soil											
P9GT09	8.27 ± 0.09	$61,000 \pm 10,000$	—	$3,200 \pm 2,200$	—	$5,120 \pm 870$	—	—	390 ± 150	92 ± 44	—
Low-Concentration Soil											
P9GT08	8.28 ± 0.02	$20,000 \pm 4,000$	670,000	$7,200 \pm 2,900$	—	$16,600 \pm 4,000$	—	770	87	47 ± 16	—
P9GT10	8.44 ± 0.10	—	—	$1,910 \pm 490$	—	$3,690 \pm 510$	$4,900 \pm 1,600$	—	—	53.4 ± 6.2	—
Mixed Soil-Waste											
P9GT03	8.73 ± 0.26	—	$350,000 \pm 110,000$	$2,700 \pm 2,400$	—	$3,400 \pm 1,500$	2,200	650	$1,930 \pm 970$	70 ± 39	—
P9GT07	8.31 ± 0.19	$27,000 \pm 25,000$	330,000	$1,400 \pm 1,100$	—	$3,700 \pm 2,900$	$9,100 \pm 2,000$	—	300	21.8 ± 4.6	—
P9GT17	9.14 ± 0.06	$6,400 \pm 1,700$	$6,900 \pm 2,400$	720 ± 190	—	$1,250 \pm 460$	$2,400 \pm 1,300$	—	75	17.3 ± 5.1	—
Soil Scraped From Graphite											
P9GT27	8.21 ± 0.08	—	—	$1,430 \pm 260$	—	$2,240 \pm 110$	$1,140,000 \pm 640,000$	930,000	$204,000 \pm 72,000$	40.7 ± 7.0	11
Soil After Rupture Of Graphite Scarfings Jar											
P9GT32	8.43 ± 0.05	$10,750 \pm 610$	$600,000 \pm 230,000$	$5,600 \pm 2,100$	640	$5,530 \pm 840$	$1,320,000 \pm 800,000$	770,000	—	43 ± 14	22
P9GT34	8.37 ± 0.14	$9,900 \pm 1,300$	—	$4,380 \pm 980$	$1,000 \pm 270$	$5,530 \pm 630$	$2,600,000 \pm 630,000$	$3,400,000 \pm 2,800,000$	—	34.8 ± 7.7	11.3 ± 5.8
Organic Waste											
P9GR04	11.81 ± 0.02	$32,500 \pm 4,400$	—	$42,100 \pm 6,400$	—	$39,600 \pm 6,200$	$310,000 \pm 170,000$	$250,000 \pm 150,000$	$270,000 \pm 110,000$	123 ± 16	36
P9GR20	9.99 ± 0.21	$41,000 \pm 11,000$	—	$20,400 \pm 10,000$	$5,580 \pm 590$	$25,000 \pm 9,500$	$79,000 \pm 52,000$	$84,000 \pm 53,000$	$96,000 \pm 64,000$	86 ± 23	13.5 ± 5.9
P9GR23	9.25 ± 0.02	$9,400 \pm 6,700$	66,000	$12,800 \pm 9,000$	—	$36,000 \pm 29,000$	33,000	$58,675.97 \pm 0.44$	—	10.1 ± 2.2	—
Overburden Soil											
P9GW09	8.47 ± 0.07	—	470,000	—	—	$9,400 \pm 2,300$	—	—	—	—	—
P9GW13	8.41 ± 0.04	—	570,000	—	—	$8,860 \pm 400$	—	—	—	—	—
P9GW15	8.54 ± 0.04	—	—	—	—	$10,400 \pm 1,500$	—	—	—	—	—
Blank INL Soil	7.89 ± 0.05	—	480,000	—	—	$7,340 \pm 510$	—	—	—	65	—
Spiked INL Soil	8.21 ± 0.04	100,000	—	604 ± 25	—	$18,500 \pm 1,900$	$34,200 \pm 1,900$	$44,300 \pm 7,700$	—	1.19 ± 0.11	122 ± 19

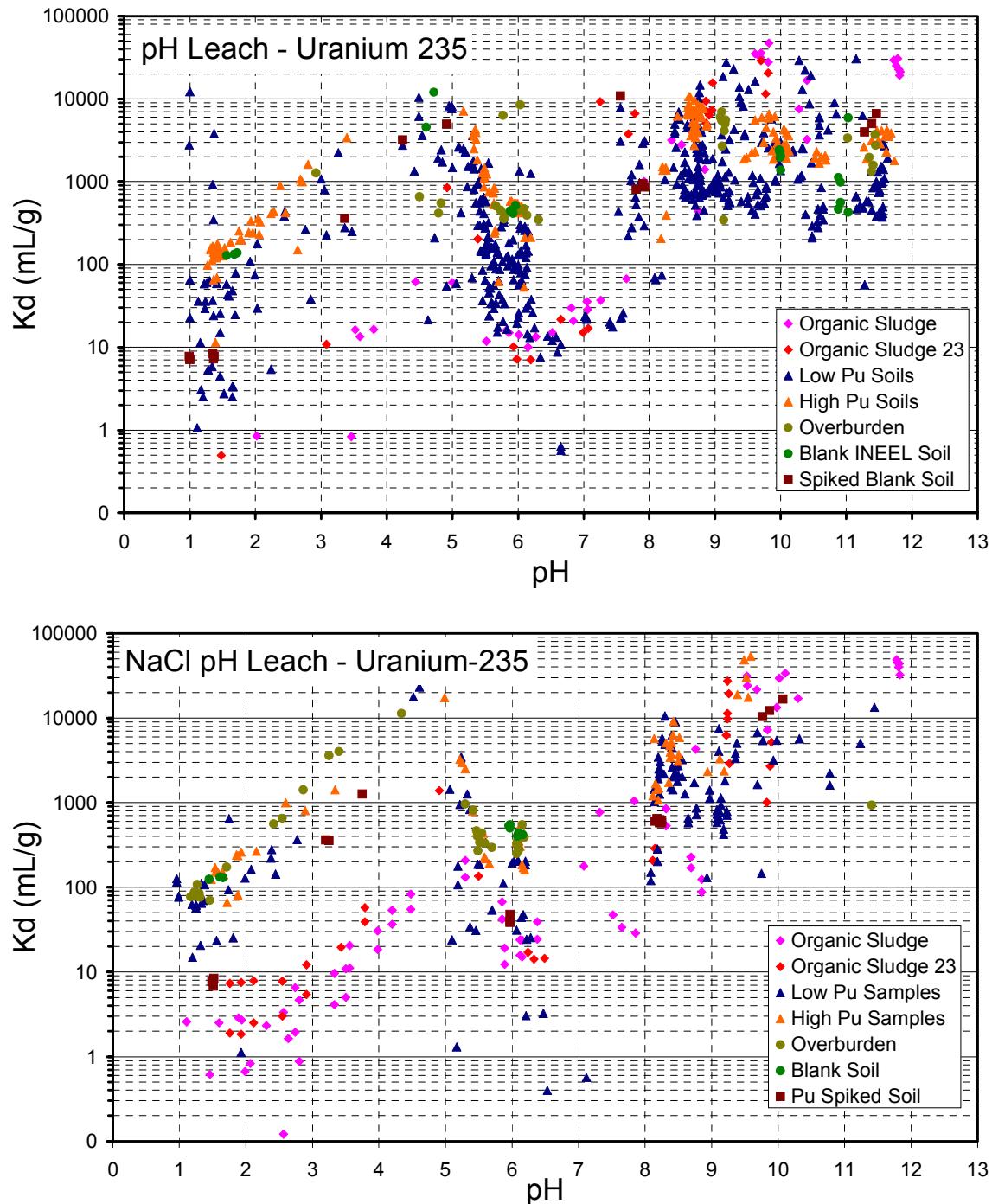


Figure F-1. Summary of K_d values for uranium at m/z 235 determined for the various interstitial soil samples, organic sludge samples, overburden soil samples, and blank and plutonium-spiked blank soil samples with deionized water (top) and a 100 mM NaCl solution (bottom) as the leachate.

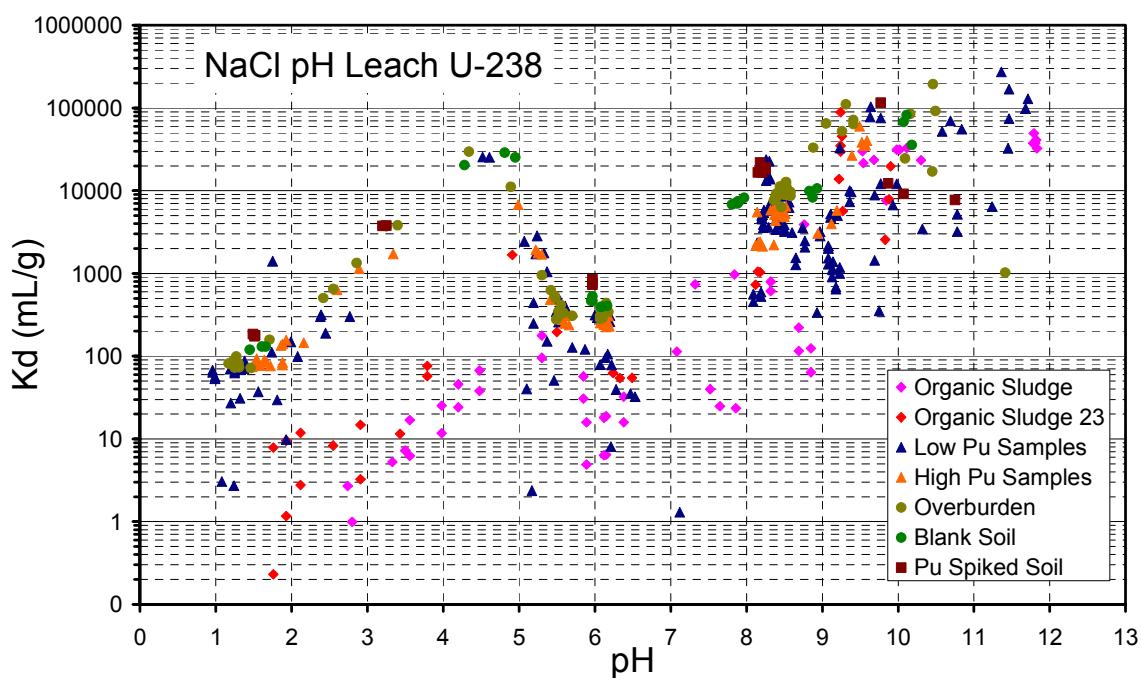
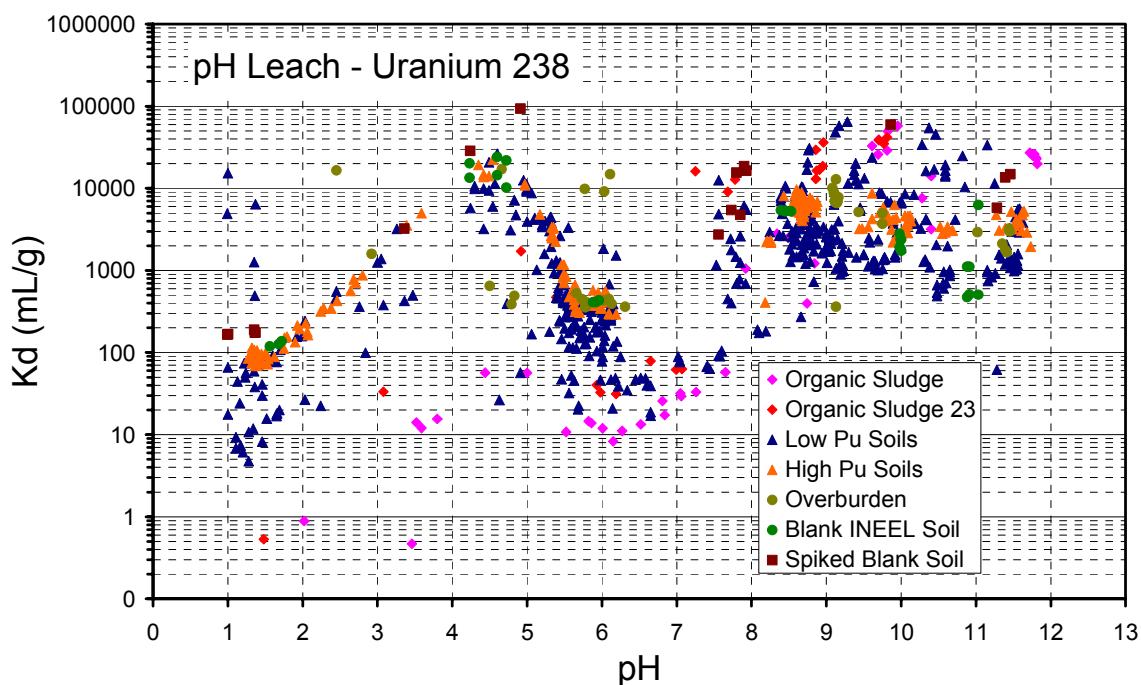


Figure F-2. Summary of K_d values for uranium at m/z 238 determined for the various interstitial soil samples, organic sludge samples, overburden soil samples, and blank and plutonium-spiked blank soil samples with deionized water (top) and a 100 mM NaCl solution (bottom) as the leachate.

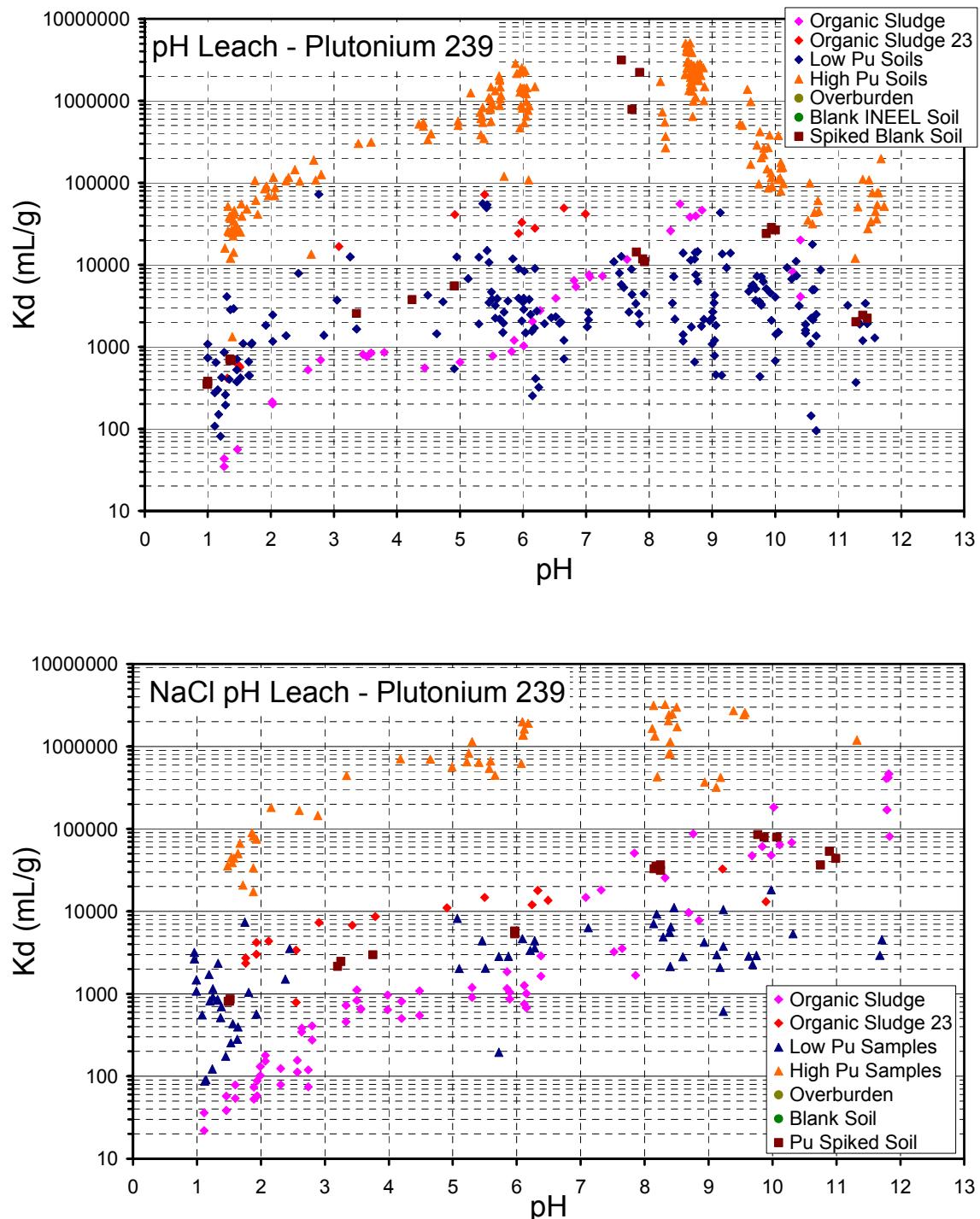


Figure F-3. Summary of K_d values for plutonium at *m/z* 239 determined for the various interstitial soil samples, organic sludge samples, overburden soil samples, and blank and plutonium-spiked blank soil samples with deionized water (top) and a 100 mM NaCl solution (bottom) as the leachate.

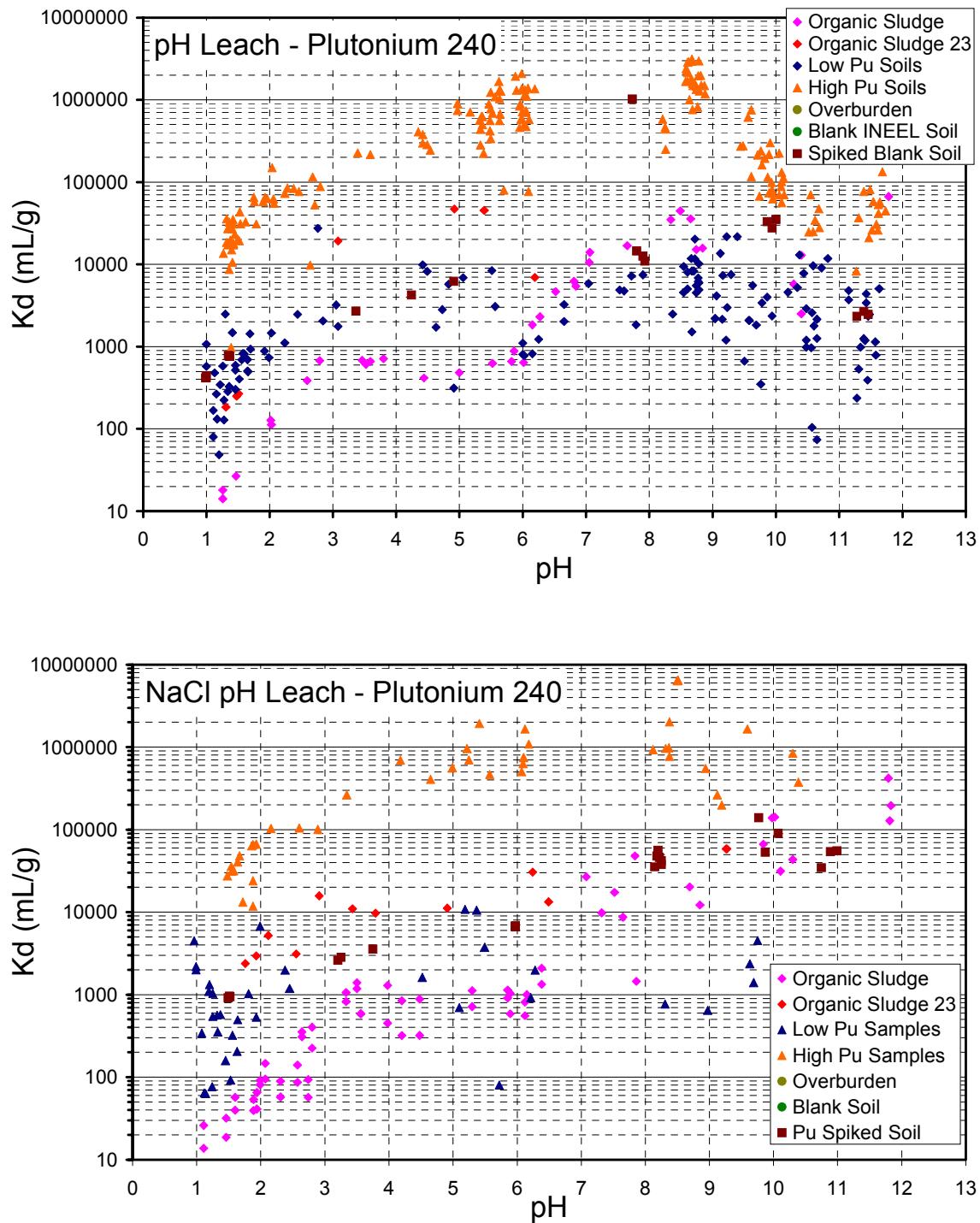


Figure F-4. Summary of Kd values for plutonium at m/z 240 determined for the various interstitial soil samples, organic sludge samples, overburden soil samples, and blank and plutonium-spiked blank soil samples with deionized water (top) and a 100 mM NaCl solution (bottom) as the leachate.

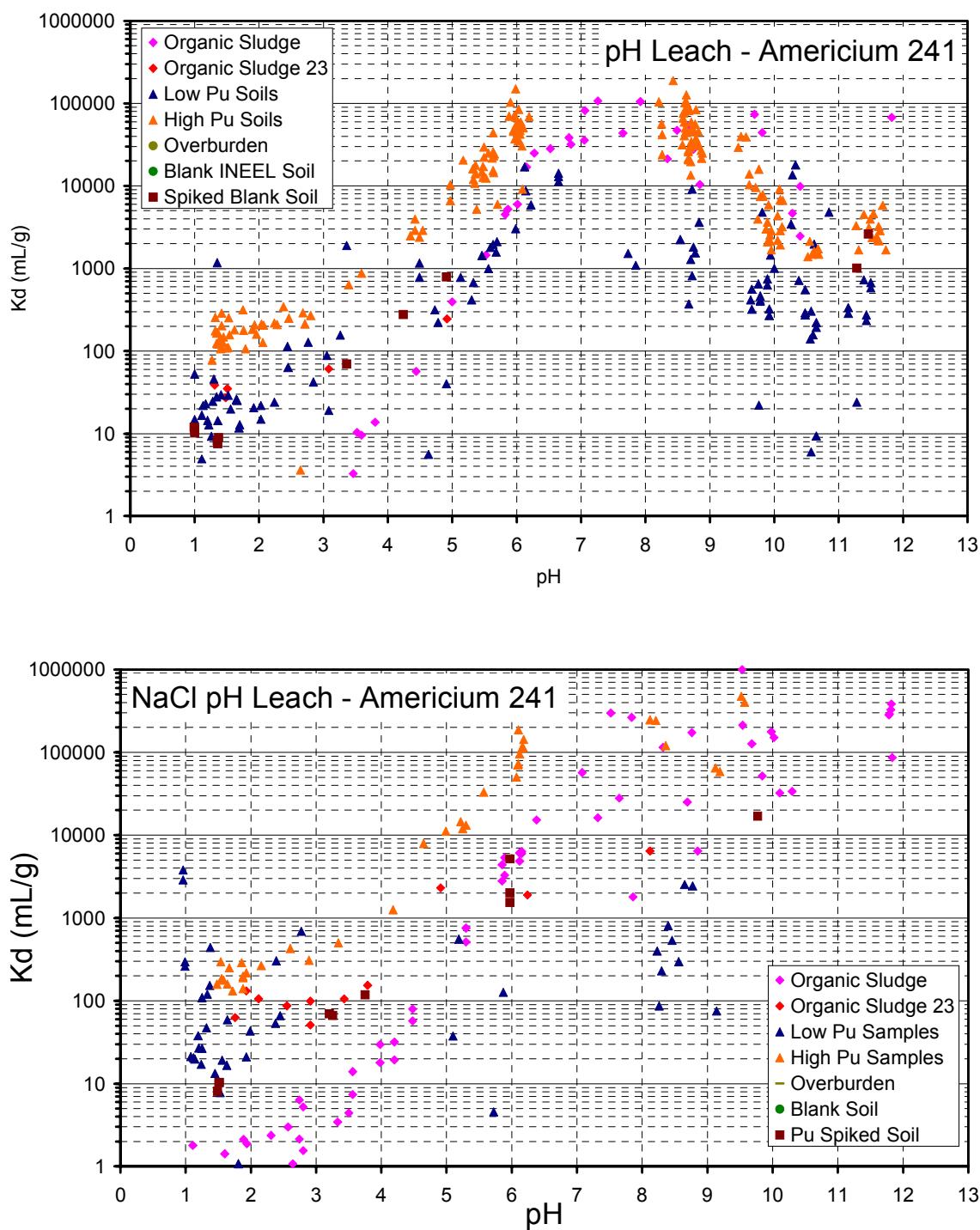


Figure F-5. Summary of K_d values for americium at *m/z* 241 determined for the various interstitial soil samples, organic sludge samples, overburden soil samples, and blank and plutonium-spiked blank soil samples with deionized water (top) and a 100 mM NaCl solution (bottom) as the leachate.

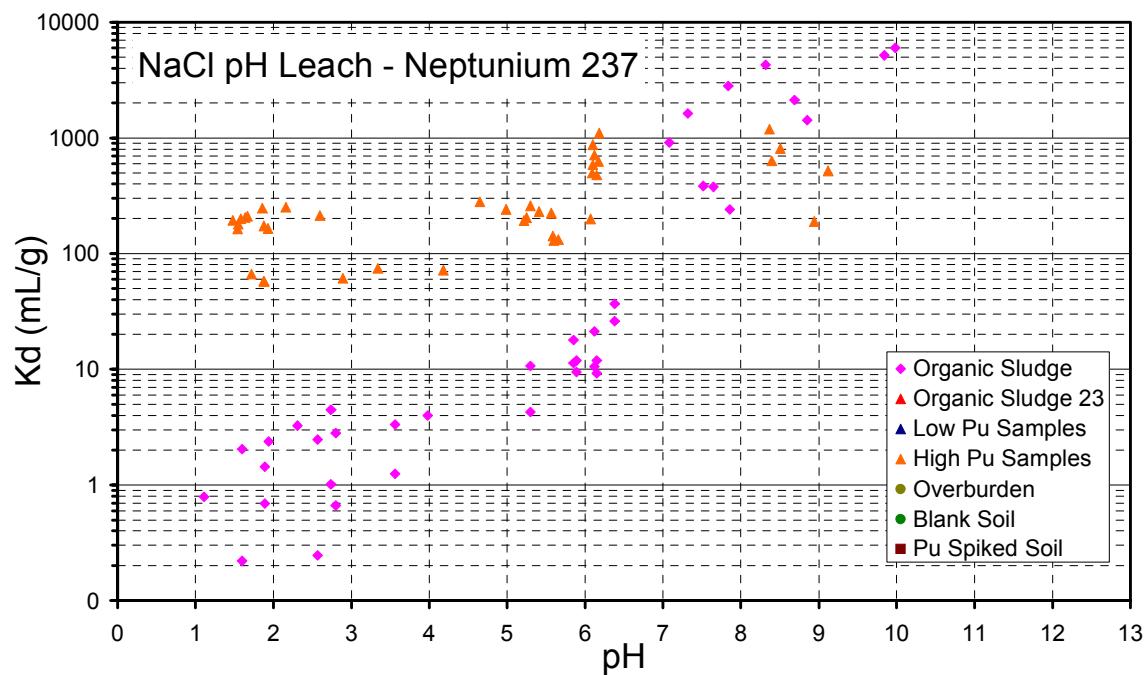
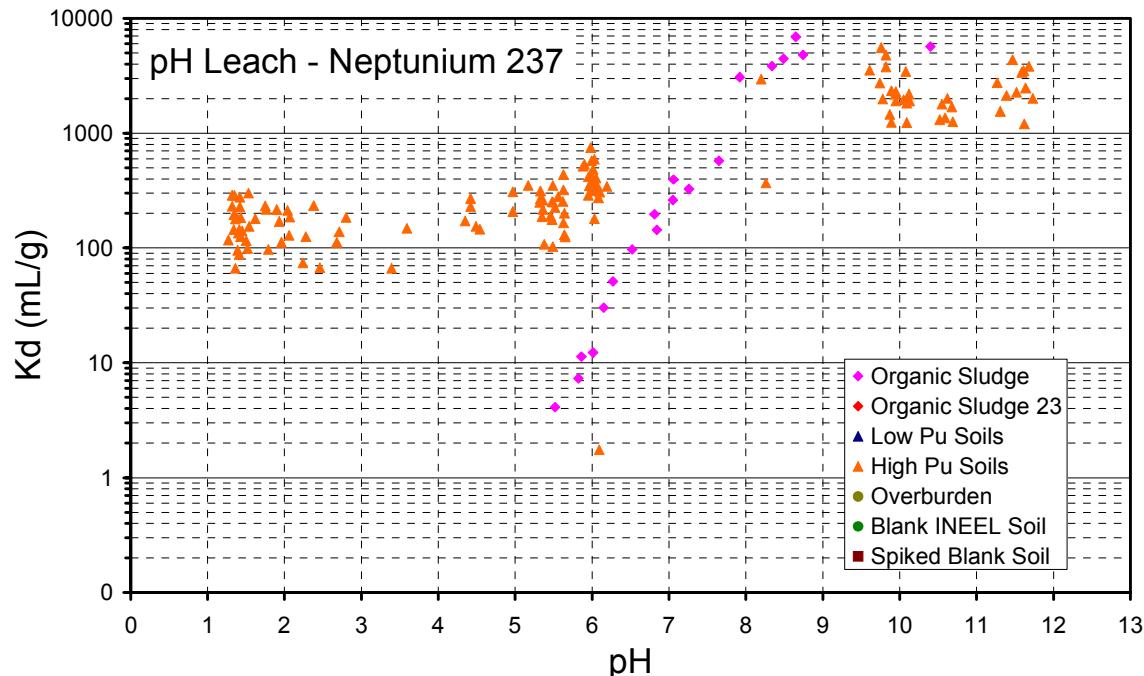


Figure F-6. Summary of K_d values for neptunium at m/z 237 determined for the various interstitial soil samples, organic sludge samples, overburden soil samples, and blank and plutonium-spiked blank soil samples with deionized water (top) and a 100 mM NaCl solution (bottom) as the leachate.

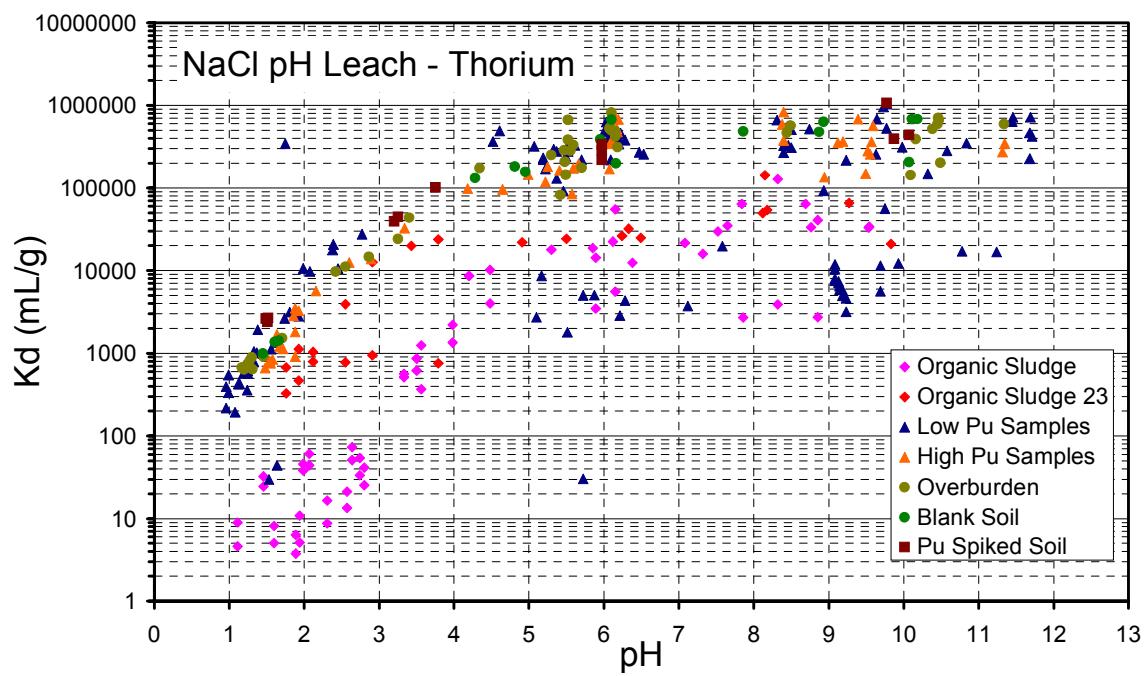
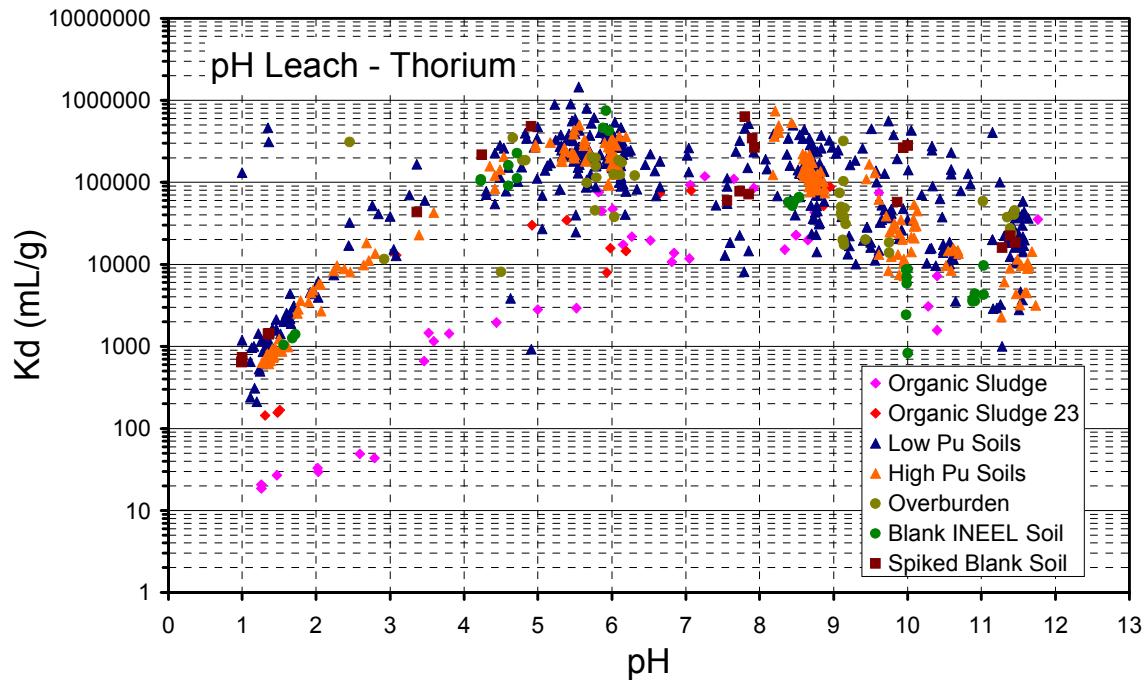


Figure F-7. Summary of K_d values for thorium determined for the various interstitial soil samples, organic sludge samples, overburden soil samples, and blank and plutonium-spiked blank soil samples with deionized water (top) and a 100 mM NaCl solution (bottom) as the leachate.

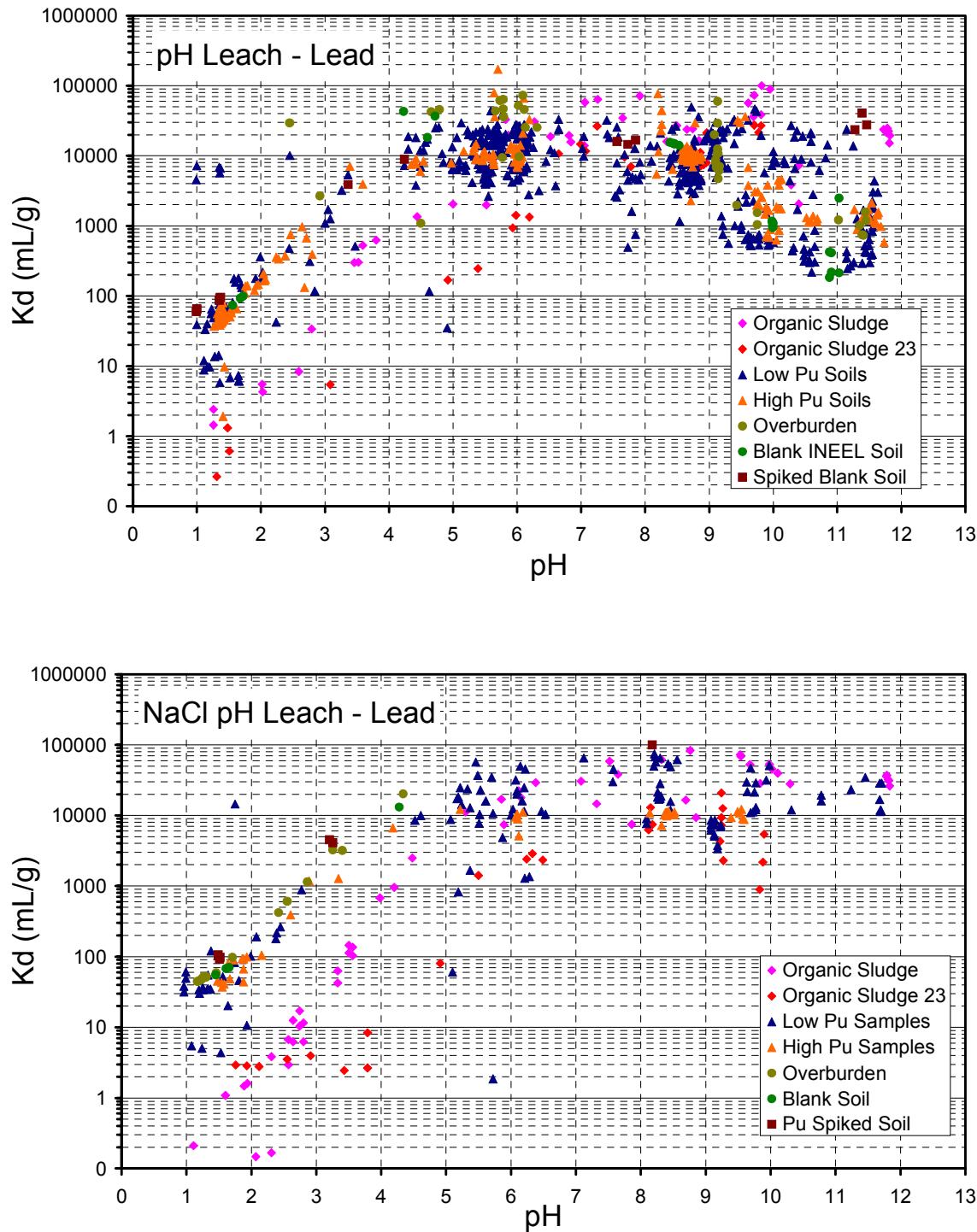


Figure F-8. Summary of K_d values for lead determined for the various interstitial soil samples, organic sludge samples, overburden soil samples, and blank and plutonium-spiked blank soil samples with deionized water (top) and a 100 mM NaCl solution (bottom) as the leachate.

Appendix G

Sequential Aqueous Extraction

Appendix G

Sequential Aqueous Extraction

The information in the following appendix summarizes the experimental procedure for the sequential aqueous extraction (SAE) experiments (see Section 9). The laboratory method determines the percentage each actinide was released in the exchangeable, carbonate, oxidizable, reducible, or residual fractions.

G-1. EXPERIMENTAL PROCEDURE FOR SEQUENTIAL AQUEOUS EXTRACTIONS

Three different aliquots of about 1 g each of soil were extracted with 10-mL of each solution in a polycarbonate, screw-capped centrifuge tube. Following contact, the sample was centrifuged, and the aqueous phase removed with a pipette, acidified, and filtered through a 0.45- μm filter. This solution was then diluted as appropriate for inductively coupled plasma-mass spectroscopy (ICP-MS) analysis. The soil was then washed with a few milliliters of deionized water, re-centrifuged, and the wash water was discarded. The soil was then subjected to the next extraction step in the sequence.

The sequence of extractions was as follows:

1. The sample was contacted for 1-hour with 10 mL of 0.01-M CaCl_2 to evaluate ion-exchangeable metals.
2. The sample was then contacted for 5 hours with 10 mL of a solution of 0.5-M sodium acetate, previously adjusted to pH 5 with acetic acid to evaluate acetate-soluble trace metals.
3. The sample was then contacted for 1 to 2-hours with 10 mL of sodium hypochlorite solution, previously adjusted to pH 9.5 with HCl, at 90–95°C. This step was repeated three times for a total final volume of 30 mL, to evaluate trace metals susceptible to dissolution in the presence of a strong oxidizing agent.
4. The sample was then contacted for 15-minutes with 10-mL of 0.3-M sodium citrate, previously buffered with 0.8-mL of 1-M sodium bicarbonate. This was performed at 80°C, and about 100 mg of sodium dithionite was added after the solution was at temperature. Then, an additional about 100 mg of sodium dithionite was added for an additional 10-minute contact at the same temperature.
5. The residual trace metal content was determined by total post-extraction soil dissolution using a sodium peroxide fusion, followed by ICP-MS analysis of the fused sample.

Mass balance calculations were performed for each sample based on total actinide concentrations previously determined for each sample by sodium peroxide fusion and ICP-MS analysis, the concentration of actinides removed from each sample during each SAE step, and the residual concentration of actinides remaining in each sample after SAE Step 4 was completed.

Table G-1. The fractional abundances of ^{238}U and ^{235}U metal in sequential aqueous extraction fractions. Uncertainty values represent one standard deviation. Column headers denote sample number and radionuclide.

Overburden			Low-Contamination Soil					
	Overburden, W09, W13, W15, ^{238}U	U from Plutonium-Spiked Soil, ^{238}U	Uncontaminated INL Soil, ^{238}U	T08, ^{238}U	T09, ^{238}U	T10, ^{238}U	—	—
Exchangeable	0.1 \pm 0.1	0.1 \pm 0.0	0.2 \pm 0.1	0.1 \pm 0.1	0.1 \pm 0.1	0.1 \pm 0.1	—	—
Carbonate	19.9 \pm 10.3	4.2 \pm 0.7	9.6 \pm 7.6	9.8 \pm 9.8	11.9 \pm 13.3	13.9 \pm 6.8	—	—
Oxidizable	18.1 \pm 10.9	5.1 \pm 0.8	10.5 \pm 8.7	10.4 \pm 9.2	49.7 \pm 27.6	22.8 \pm 12.4	—	—
Reducible	14.2 \pm 7.2	8.8 \pm 1.7	4.0 \pm 4.3	3.4 \pm 3.2	6.2 \pm 7.2	3.1 \pm 1.9	—	—
Residual	48.0 \pm 27.5	81.7 \pm 3.1	75.8 \pm 20.7	76.1 \pm 22.1	32.1 \pm 29.7	60.2 \pm 18.1	—	—
Mixed Soil-Waste								
	T03, ^{238}U	T03, ^{235}U	T05, ^{238}U	T05, ^{235}U	T07, ^{238}U	T07, ^{235}U	T17, ^{238}U	T17, ^{235}U
Exchangeable	0.2 \pm 0.0	4.2 \pm 1.6	0.1 \pm 0.1	0.0 \pm 0.0	1.1 \pm 0.9	1.2 \pm 1.6	0.0 \pm 0.0	0.0 \pm 0.0
Carbonate	23.4 \pm 10.4	36.2 \pm 12.1	25.2 \pm 11.3	5.0 \pm 0.4	12.4 \pm 4.4	22.3 \pm 4.2	5.4 \pm 0.8	11.6 \pm 1.7
Oxidizable	39.8 \pm 13.5	19.7 \pm 18.6	24.4 \pm 11.5	65.5 \pm 4.4	49.4 \pm 25.9	36.0 \pm 26.4	1.0 \pm 0.2	1.6 \pm 0.8
Reducible	6.1 \pm 1.9	5.7 \pm 2.2	9.4 \pm 6.5	21.3 \pm 4.8	20.0 \pm 3.9	20.1 \pm 1.4	1.5 \pm 0.3	2.9 \pm 1.0
Residual	30.5 \pm 24.5	34.2 \pm 27.0	40.9 \pm 29.1	8.3 \pm 6.6	17.1 \pm 27.8	30.5 \pm 36.8	92.1 \pm 1.3	83.8 \pm 3.4
Soil Scraped from Graphite, Soil After Rupture of Graphite Scarfings Jar								
	T27, ^{238}U	T27, ^{235}U	T32, ^{238}U	T32, ^{235}U	T34, ^{238}U	T34, ^{235}U	—	—
Exchangeable	1.1 \pm 1.6	0.0 \pm 0.0	0.1 \pm 0.2	0.0 \pm 0.0	0.1 \pm 0.0	0.0 \pm 0.0	—	—
Carbonate	6.0 \pm 2.5	8.7 \pm 3.5	3.0 \pm 2.5	2.5 \pm 2.0	2.7 \pm 0.3	32.4 \pm 13.8	—	—
Oxidizable	7.5 \pm 2.6	18.8 \pm 5.5	3.7 \pm 2.6	4.4 \pm 3.1	6.5 \pm 1.5	5.8 \pm 1.8	—	—
Reducible	3.1 \pm 3.1	6.0 \pm 0.8	2.2 \pm 1.6	1.4 \pm 1.0	4.3 \pm 0.8	1.9 \pm 0.6	—	—
Residual	83.3 \pm 8.2	66.4 \pm 7.7	90.9 \pm 6.8	91.9 \pm 5.7	86.3 \pm 2.5	59.9 \pm 16.1	—	—

	Organic Waste					
	R04, ^{238}U	R04, ^{235}U	R20, ^{238}U	R20, ^{235}U	R23, ^{238}U	R23, ^{235}U
Exchangeable	0.0 ± 0.0	—				
Carbonate	7.5 ± 5.6	7.8 ± 5.4	39.9 ± 9.4	38.9 ± 7.8	6.1 ± 0.3	4.9 ± 5.8
Oxidizable	10.7 ± 2.1	13.2 ± 1.9	33.3 ± 8.9	29.8 ± 8.8	0.4 ± 0.2	35.1 ± 36.1
Reducible	7.7 ± 3.2	7.8 ± 2.5	3.7 ± 0.9	3.7 ± 0.9	1.3 ± 0.3	5.0 ± 3.0
Residual	74.1 ± 8.2	71.2 ± 6.0	23.1 ± 17.5	27.4 ± 16.1	92.2 ± 0.8	55.0 ± 31.3

Table G-2. the fractional abundances of plutonium metal in sequential aqueous extraction fractions. Uncertainty values represent one standard deviation.

Low-Contamination Soil				
	Plutonium-spiked soils	T08 Plutonium	T09 Plutonium	T10 Plutonium
Exchangeable	0.1 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Carbonate	0.0 ± 0.0	2.4 ± 4.1	0.0 ± 0.0	3.0 ± 2.0
Oxidizable	73.3 ± 1.7	51.4 ± 30.4	76.3 ± 13.9	38.4 ± 13.6
Reducible	24.5 ± 1.5	21.0 ± 12.1	21.8 ± 11.5	27.8 ± 10.5
Residual	2.4 ± 1.1	25.2 ± 22.0	2.0 ± 3.5	30.9 ± 24.7
Mixed Soil-Waste				
	T03 Plutonium	T05 Plutonium	T07 Plutonium	T17 Plutonium
Exchangeable	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Carbonate	0.0 ± 0.0	0.8 ± 0.7	2.9 ± 2.1	0.4 ± 0.8
Oxidizable	57.5 ± 27.7	8.1 ± 3.4	24.5 ± 16.2	7.3 ± 6.6
Reducible	27.9 ± 11.6	90.0 ± 4.7	35.3 ± 15.2	12.7 ± 3.9
Residual	14.7 ± 16.4	0.0 ± 0.0	55.9 ± 3.4	80.6 ± 9.0
Soil Scraped from Graphite, Soil After Rupture of Graphite Scarfings Jar				
	T27 Plutonium	T32 Plutonium	T34 Plutonium	
Exchangeable	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	—
Carbonate	0.1 ± 0.0	0.1 ± 0.2	3.5 ± 4.1	—
Oxidizable	34.9 ± 4.2	16.3 ± 19.7	4.0 ± 0.8	—
Reducible	12.5 ± 1.1	10.9 ± 13.6	2.7 ± 0.5	—
Residual	52.5 ± 3.5	71.2 ± 35.5	87.7 ± 6.5	—
Organic Waste				
	Plutonium-spiked soils	R04, Plutonium	R20, Plutonium	R23, Plutonium
Exchangeable	0.1 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Carbonate	0.0 ± 0.0	0.5 ± 0.6	0.5 ± 0.6	1.4 ± 0.1
Oxidizable	73.3 ± 1.7	8.2 ± 1.4	8.2 ± 1.4	1.7 ± 0.9
Reducible	24.5 ± 1.5	6.8 ± 0.2	6.8 ± 0.2	26.5 ± 10.0
Residual	2.4 ± 1.1	85.0 ± 1.2	85.0 ± 1.2	71.9 ± 11.3

Table G-3. The fractional abundances of americium metal in sequential aqueous extraction fractions. Uncertainty values represent one standard deviation.

Soil Scrapped from Graphite, Soil After Rupture of Graphite Scarfings Jar			
	T27 Americium	T32 Americium	T34 Americium
Exchangeable	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Carbonate	2.2 ± 0.6	0.9 ± 0.7	1.4 ± 1.0
Oxidizable	15.6 ± 1.6	2.8 ± 1.6	2.9 ± 0.8
Reducible	18.6 ± 2.4	2.3 ± 0.4	4.3 ± 2.0
Residual	63.7 ± 0.6	91.5 ± 5.9	92.3 ± 1.7
Organic Waste			
	R04 Americium	R20 Americium	R23 Americium
Exchangeable	0.0 ± 0.0	0.0 ± 0.0	—
Carbonate	0.3 ± 0.5	2.9 ± 0.6	—
Oxidizable	1.0 ± 0.3	1.6 ± 0.5	—
Reducible	13.4 ± 8.6	20.4 ± 13.7	—
Residual	88.1 ± 2.4	75.1 ± 13.7	—

Table G-4. The fractional abundances of neptunium metal in sequential aqueous extraction fractions. Uncertainty values represent one standard deviation.

Soil Scraped from Graphite, Soil After Rupture of Graphite Scarfings Jar			
	T27 Neptunium		
Exchangeable	0.0 ± 0.0	—	—
Carbonate	26.9 ± 10.3	—	—
Oxidizable	73.1 ± 10.3	—	—
Reducible	0.0 ± 0.0	—	—
Residual	0.0 ± 0.0	—	—
Organic Waste			
	R04 Neptunium	R20 Neptunium	
Exchangeable	0.0 ± 0.0	0.0 ± 0.0	—
Carbonate	7.2 ± 6.0	20.5 ± 16.3	—
Oxidizable	10.8 ± 6.3	67.7 ± 21.7	—
Reducible	28.3 ± 32.3	4.3 ± 2.0	—
Residual	53.8 ± 26.0	7.5 ± 4.8	—

Appendix H

Surface Characterization Using Ion Trap Secondary Ion Mass Spectrometry

Appendix H

Surface Characterization Using Ion Trap Secondary Ion Mass Spectrometry

H-1. EXPERIMENTAL DETAILS OF SIMS CHARACTERIZATION

Ion trap-secondary ion mass spectrometry (IT-SIMS) was used to provide a qualitative evaluation of the surface chemistry of the soil and waste samples. IT-SIMS functions by bombarding samples with energetic molecules, which causes chemicals on the surface to be sputtered into the gas phase of an ion trap mass spectrometer. Some of the chemicals produced by the bombardment process will have a positive or negative charge, which enables mass measurement.

The spectra were obtained using a modified Varian Saturn 2,000 ion trap mass spectrometer (Walnut Creek, CA) adapted for secondary ion mass spectrometry. Briefly, the instrument is equipped with a ReO_4^- primary ion gun, an offset venetian-blind dynode/multichannel plate detector system housed in a custom-fabricated vacuum chamber. The ReO_4^- primary particle provides enhanced production of molecular secondary ions compared to atomic projectiles.

The ReO_4^- ion gun is mounted coaxial with axis of the ion trap and the beam enters the ion trap through an aperture in the top end cap, passes along the central axis of the ion trap mass analyzer, and strikes the sample located behind the opposite end cap. The ReO_4^- ion gun was operated at 5.0 keV, at a primary ion current of 700 to 800 picoamps (measurable using a retractable Faraday cup). Secondary ions sputtered from the sample surface are focused into the ion trap by a small cylindrical electrostatic lens, where upon entering the trap, the secondary ions undergo collisions with the He bath gas (3×10^{-5} torr), and lose sufficient kinetic energy such that they are trapped by the oscillating RF field. Full scan cation and anion spectra were obtained for each of the sample at base mass cutoffs of 15 and 40 amu and 20 and 40 amu, respectively.

Samples are prepared for SIMS analysis by attaching 2–4 mg of powder or particulate to an insertion probe using double sided adhesive tape. The sample is not altered in any other way, and the surface facing the bombarding beam (i.e., not in contact with the tape) has not been modified, and hence reflects the chemistry of the top-most molecular layers of the sample.

H-2. DEVELOPMENT OF EMPIRICAL METHOD FOR THE DETERMINATION OF WASTE FORM TYPE

The purpose of this effort was to utilize IT-SIMS to categorize soil and waste samples on the basis of their surface chemistry. Comparison of spectra of soil and putative waste samples suggested that an empirical differentiation could be generated, which would have significant utility for categorizing highly radioactive samples whose handling is limited to microgram sizes to minimize dose. Differentiation in the cation spectra from organic wastes and soils could be achieved by differences in the abundances of the “envelopes” of hydrocarbon peaks (Figure H-1). Furthermore, a bias of the C_7 envelope was noted in the $m/z\ 91^+/\text{C}_7^+$ ion ratio: the value was reproducibly smaller in the waste samples compared to the soils. These differences formed the basis for the development of an empirical method for the distinguishing sample-types. Three determinate parameters were identified for differentiation:

1. The ratio of $m/z\ 91^+$ to $m/z\ 95^+$;

2. The ratio of the C₅ envelope (*m/z* 67, 69, 71, 73) abundance (most abundant ion of the series) to the base peak abundance; and
3. The ratio of the C₆ envelope (*m/z* 79, 81, 83, 85) abundance (most abundant ion of the series) to the base peak abundance of the cation spectra. The empirical data derived from the benchmark soil and the organic waste samples was compiled in Table H-1.

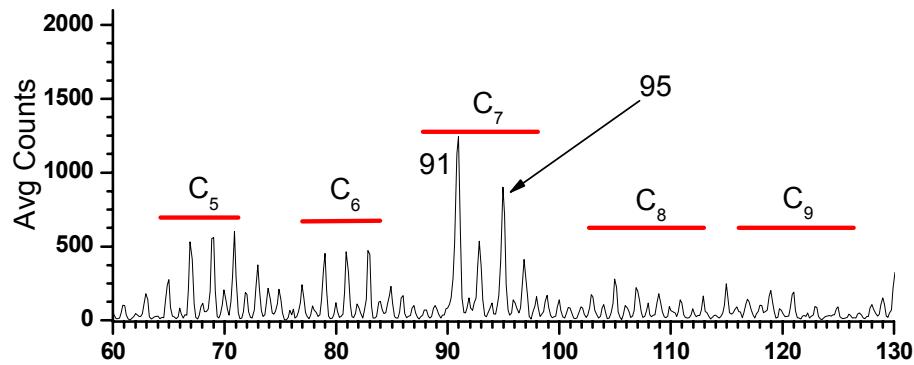


Figure H-1. Hydrocarbon ion envelopes for the soil sample in the IT-SIMS spectrum.

Table H-1. Values of determinate parameters for sample differentiation based on ion abundances from the SIMS spectra.

Determinate Samples	67-73 ⁺ /BP Ion Ratio	79-85 ⁺ /BP Ion Ratio	91/95 ⁺ Ion Ratio	62/60 ⁺ Ion Ratio	Anion Spectrum
Benchmark Overburden Soil Samples					
W04	0.35	0.17	1.97	0.16	good
W09	0.24	0.16	1.07	0.06	good
W12	0.09	0.14	3.27	0.11	good
W13	0.13	0.11	1.28	0.18	good
W15	0.32	0.27	1.36	0.06	good
W21	0.50	0.51	1.37	0.10	good
Average	0.27	0.23	1.72	0.11	—
Std Dev (<i>s</i>)	0.15	0.15	0.82	0.05	—
Upper <i>s</i> Limit	0.42	0.38	2.54	—	—
Lower <i>s</i> Limit	0.12	0.08	0.90	—	—
Accepted Organic Sludge					
R20	0.73	0.59	0.98	NA	poor
R23	1.00	0.86	0.70	NA	poor
Average	0.87	0.73	0.84	—	—
Std Dev (<i>s</i>)	0.19	0.19	0.20	—	—
Upper <i>s</i> Limit	1.06	0.92	1.04	—	—
Lower <i>s</i> Limit	0.67	0.53	0.64	—	—
P9GR04012G	0.19	0.24	0.91	10.3	—

Table H-1. (continued).

Determinate Samples	67-73 ⁺ /BP Ion Ratio	79-85 ⁺ /BP Ion Ratio	91/95 ⁺ Ion Ratio	62/60 ⁺ Ion Ratio	Anion Spectrum
Unknown Waste Form					
P01	0.53	0.38	1.70	0.44	good
P02	NA	NA	NA	1.19	good
P03	0.31	0.18	1.69	2.57	good
P04	0.32	0.15	2.95	0.80	good
P05	0.27	0.27	2.04	1.84	good
Average				1.37	
Std Dev (s)				0.85	
Upper s Limit				2.22	
Lower s Limit				0.52	
BP = Base Peak NA = data not available					

For the benchmark overburden soil samples the 91⁺/95⁺ ratio average (\tilde{x}) is 1.72 with standard deviation (s) of 0.82. The upper s value ($\tilde{x} + s$) is 2.54 and the lower s value is 0.90. The 67–73⁺ ion series/base peak ratio average is 0.27 with $s = 0.15$. The upper s value is 0.42. The 79–85⁺ ion series/base peak ratio average is 0.23 with $s = 0.15$ and the upper s value = 0.38. For discussion the 91⁺/95⁺ ratio is considered the primary ratio, with the 67–73⁺ ion series/base peak ratio and the 79–85⁺ ion series/base peak ratio considered as secondary ratios.

For the organic waste samples the 91⁺/95⁺ ratio average is 0.84 with $s = 0.20$. The upper s value is 1.04 and lower s value is 0.64. The 67–73⁺ ion series/base peak ratio average is 0.87 with $s = 0.19$. The upper s value is 1.06, whereas the lower s value is 0.67. The 79–85⁺ ion series/base peak ratio average is 0.73 with $s = 0.19$. The upper s value is 0.92 and the lower s value is 0.53. For the accepted inorganic sludges the 62/60⁺ ratio average is 1.37 with $s = 0.85$. The upper s limit is 2.22.

A review of the spectral data showed that many of the interstitial samples had a strong hydrocarbon appearance; the cation spectrum of mixed soil-waste sample T19 is exemplary (Figure H-2). Normally, the anion spectra contained complementary information providing some indication of an inorganic nature of the samples. These considerations enabled the development of four determinate parameters for *categories* of samples that were then used to characterize the interstitial samples (Table H-2).

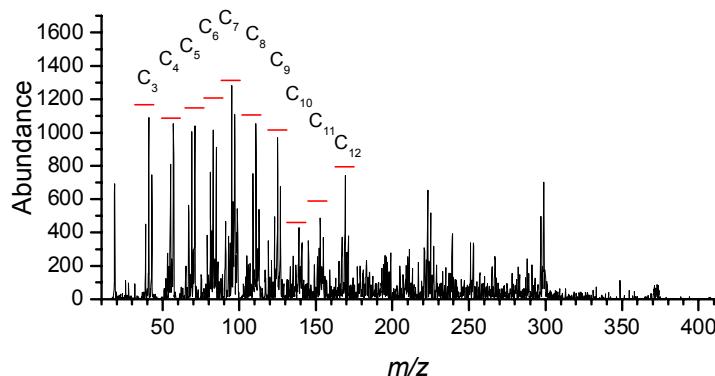


Figure H-2. Cation spectrum of mixed soil-waste sample T19, showing very prominent hydrocarbon envelopes typical of organic contamination.

Table H-2. Categories of samples derived from the determinate parameters in Table H-1.

Identified Waste Form	91 ⁺ /95 ⁺ Ion Ratio	67–73 ⁺ /BP Ion Ratio	79–85 ⁺ /BP Ion Ratio	60 ⁻ /62 ⁻ Ion Ratio
Soil	≥ 2.54			
Soil	$1.72 \leq r < 2.54$	and	< 0.42	or
Primarily Soil	< 1.72	and	< 0.42	and
Sludge/Soil Mixture	$0.90 \leq r \leq 1.04$			
Primarily Organic	≤ 0.90	and	< 0.67	or
Organic Sludge	$0.64 < r \leq 0.84$	and	> 0.67	or
Organic Sludge	≤ 0.64			
Unknown Waste Forms				≥ 2.22

H-3. RESULTS

Categorization of the 36 soil samples, using the determinate parameters in Table H-1, resulted in an independent categorization: 15 were characterized as soil samples (clean), nine were characterized as organic sludges, eight were characterized as primarily soil (clean to mostly clean), one was characterized as primarily organic sludge, and three were characterized as soil /sludge mix (Table H-3). One caveat for this categorization was that the ion counts (anions) for several samples were particularly low (designated NA in Table H-3), which may bias the ion ratios and distort the results. Nevertheless, agreement with categorization by appearance was deemed good. The results suggest that with refinement the technique could provide substantial information on sample category and much chemical information.

Table H-3. Categorization of samples using the determinate parameters in Table H-1 and the categories derived in Table H-2.

Interstitial Samples	67–73+/BP Ion Ratio	79–85+/BP Ion Ratio	91/95 ⁺ Ion Ratio	62/60 ⁻ Ion Ratio	Comments	Result
T01	0.44	0.20	2.54	0.23	BP ⁻ =76(500)	soil
T02	0.20	0.15	4.16	0.59	BP ⁻ =155(1,100)	soil
T03	0.96	0.94	0.55	NA	BP ⁻ =155(100)	organic sludge
T04	1.06	1.04	0.69	4.38	BP ⁻ =62(350); <i>m/z</i> 60 ⁻ (80)	in/organic sludge
T05	0.23	0.20	1.34	1.17	BP ⁻ =95(160)	primarily soil
T06	0.85	0.53	0.56	2.60	BP ⁻ =95(140)	in/organic sludge
T07	0.55	0.48	0.80	NA	BP ⁻ =155(120)	primarily organic
T08	0.24	0.16	2.41	0.09	BP ⁻ =95(820)	soil
T09	0.24	0.25	1.75	0.20	BP ⁻ =95(790)	soil
T10	0.26	0.18	1.22	0.08	BP ⁻ =95(3,100)	primarily soil
T11	0.16	0.29	2.40	0.32	BP ⁻ =76(820)	soil
T12	0.17	0.15	2.99	0.25	BP ⁻ =155(550)	soil
T13	0.28	0.23	2.78	0.25	BP ⁻ =76(130)	soil
T14	0.55	0.51	0.58	NA	BP ⁻ =60(80)	organic sludge
T15	0.74	0.29	0.83	0.45	bm ⁻ =40; BP ⁻ =155(540)	organic sludge
T16	0.85	1.00	0.55	1.11	BP ⁻ =95(350)	organic sludge
T17	0.64	0.62	0.57	NA	BP ⁻ =60(100)	organic sludge
T18	0.47	0.61	0.79	0.23	BP ⁻ =155(1,490)	organic sludge
T19	0.81	0.79	0.34	NA	BP ⁻ =167(280)	organic sludge
T20	0.54	0.31	2.37	0.54	BP ⁻ =97(280)	soil
T21	0.32	0.55	1.94	0.27	BP ⁻ =155(580)	soil
T22	0.30	0.28	3.09	0.19	BP ⁻ =95(1,550)	soil
T23	0.08	0.20	2.28	0.15	BP ⁻ =155(1,130)	soil
T24	0.37	0.35	1.90	0.15	BP ⁻ =155(510)	soil
T25	0.23	0.21	1.06	0.17	<i>m/z</i> 62 ⁻ (60)	primarily soil
T26	0.22	0.29	1.25	0.11	BP ⁻ =155(1,650)	primarily soil
T27	0.26	0.12	1.76	0.24	BP ⁻ =155(950)	soil
T28	0.32	0.18	1.04	0.33	BP ⁻ =76(180); <i>m/z</i> 62 ⁻ (50)	mixture
T29	0.38	0.38	0.93	0.28	BP ⁻ =155(1,400)	mixture
T30	0.24	0.23	0.97	0.12	BP ⁻ =76(2,000)	mixture
T31	0.25	0.12	1.10	0.06	<i>m/z</i> 62 ⁻ (20)	primarily soil
T32	0.15	0.09	1.60	0.04	<i>m/z</i> 62 ⁻ (50)	primarily soil
T33	0.14	0.16	1.49	0.13	BP ⁻ =762(2,400)	primarily soil
T34	NA	NA	2.52	0.12	bm ⁺ =40; <i>m/z</i> 62 ⁻ (50)	soil
T35	0.32	0.14	1.67	0.13	<i>m/z</i> 62 ⁻ (40)	primarily soil
T36	0.09	0.18	2.00	0.29	BP+=19; <i>m/z</i> 62 ⁻ (50)	soil

BP = Base Peak

bm = base mass (amu)

NA = data not available

Appendix I

Glovebox Excavator Method Project Sample Repackaging

Appendix I

Glovebox Excavator Method Project Sample Repackaging

Table I-1, I-2 and I-3 identify the three drum identification numbers used for repackaging the unused OU 7-13/14 Glovebox Excavator Method Project Retrieved Waste Sample Characterization samples for future retrieval purposes (i.e., Bauer and Barnes NABIR 2005 Proposal). There are three 10-gal DOT Type A 7A drums (UN 1A2/Y1.2/100/04/USA/M4035; UN 1A2/X100/S/04/USA/M4035; QA# 105395) containing double-bagged Glovebox Excavator Method samples placed in one 30-gal drum currently stored in Building 661 at the Reactor Technologies Complex. The tables list the sample identity, the container type, and the approximate amount of sample contents placed in the drum. The unused sample material was a remnant of the original sample retrieved during the Glovebox Excavator Method Project for RWSC analysis. The pucks were generated after gamma spectroscopy, the Petri dishes were generated after sample photography and sample analyses. The squat jars are the remainder of the unused sample material not used in any previous analysis. All three sample containers are placed in the three individual 10-gal drums placed in the single 30-gal drum for future analyses. Table I-4 lists the isotopes present in the drum and the approximate amounts.

I-1. 10-gal Drum #1

The total weight of Drum #1 was 25 lb, net weight 7.3 lb, net volume about 5 gal., <0.5 mR/hr @ contact. The contents are three overburden soil puck samples, 36 interstitial soil puck samples, three Series 743 sludge puck samples, 35 interstitial soil Petri dish samples, two Series 743 sludge Petri dish samples, and four Series 741/742 sludge Petri dish samples.

Table I-1. Drum identification (#1) and associated sample identity contents and approximate amount of sample for each drum repackaged for future retrieval purposes.

Glovebox Excavator Method Sample Repackaging		
Drum Identification: 10-gal Drum #1		Bag JAN 4-05#1
Sample Identity	Sample Container	Approx. Amount
P9GW09013A	Puck	66g
P9GW15013A	Puck	58g
P9GW13013A	Puck	61g
P9GT03016G	Puck	68g
P9GR23012G	Puck	62g
P9GT04016G	Puck	63g
P9GT08016G	Puck	65g
P9GT11016G	Puck	64g
P9GT13016G	Puck	63g
P9GT07016G	Puck	70g
P9GT10016G	Puck	63g
P9GR20012G	Puck	65g

Table I-1. (continued).

Glovebox Excavator Method Sample Repackaging		
P9GR17012G	Puck	64g
P9GT14016G	Puck	67g
P9GR04012G	Puck	65g
P9GT05016G	Puck	58g
P9GT19016G	Puck	51g
P9GT12016G	Puck	61g
P9GT18016G	Puck	67g
P9GT16016G	Puck	57g
P9GT06016G	Puck	66g
P9GT09016G	Puck	70g
P9GT02016G	Puck	56g
P9GT15016G	Puck	63g
P9GT01016G	Puck	60g

Drum Identification 10-gal Drum #1		Bag JAN 4-05 #2
Sample Identity	Sample Cont.	Approx. Amount
P9GT05016G	Petri dish	<5 g
P9GT03016G	Petri dish	<5 g
P9GR23012G	Petri dish	10-20 g
P9GT10016G	Petri dish	5-10g
P9GT08016G	Petri dish	5 g
P9GR20012G	Petri dish	10-20 g
P9GT18016G	Petri dish	5 g
P9GT13016G	Petri dish	5 g
P9GT14016G	Petri dish	5 g
P9GT15016G	Petri dish	5-10 g
P9GT16016G	Petri dish	5-10 g
P9GT17016G	Petri dish	5-10 g
P9GT28016G	Petri dish	10-20 g
P9GT26016G	Petri dish	10-20 g
P9GT27016G	Petri dish	10-20 g
P9GT12016G	Petri dish	5-10 g
P9GT24016G	Petri dish	10-20 g
P9GT11016G	Petri dish	10-20 g
P9GT22016G	Petri dish	10-20 g

Table I-1. (continued).

Drum Identification 10-gal Drum #1		Bag JAN 4-05 #2
Sample Identity	Sample Cont.	Approx. Amount
P9GT19016G	Petri dish	5–10 g
P9GT31016G	Petri dish	10–20 g
P9GT33016G	Petri dish	10–20 g
P9GT35016G	Petri dish	10–20 g
P9GT21016G	Puck	70g
P9GT23016G	Puck	70g
P9GT29016G	Puck	70g
P9GT30016G	Puck	70g
P9GT33016G	Puck	70g
P9GT25016G	Puck	70g
P9GT20016G	Puck	70g
P9GT36016G	Puck	70g
P9GT35016G	Puck	70g
P9GT31016G	Puck	70g
P9GT22016G	Puck	70g
P9GT27016G	Puck	70g
P9GT28016G	Puck	70g
P9GT32016G	Puck	70g
P9GT24016G	Puck	70g
P9GT34016G	Puck	70g
P9GT26016G	Puck	70g
P9GT29016G	Petri dish	Variable
P9GP01015G	Petri dish	Variable
P9GT04016G	Petri dish	Variable
P9GP01015G	Petri dish	Variable
P9GP03015G	Petri dish	Variable
P9GP03015G	Petri dish	Variable
P9GT02016G	Petri dish	Variable
P9GT23016G	Petri dish	Variable
P9GT21016G	Petri dish	Variable
P9GT34016G	Petri dish	Variable
P9GP05015G	Petri dish	Variable
P9GT30016G	Petri dish	Variable

Table I-1. (continued).

Drum Identification 10-gal Drum #1		Bag JAN 4-05 #2
Sample Identity	Sample Cont.	Approx. Amount
P9GT07016G	Petri dish	Variable
P9GT07016G	Petri dish	Variable
P9GT25016G	Petri dish	Variable
P9GT36016G	Petri dish	Variable
P9GT01016G	Petri dish	Variable
P9GT06016G	Petri dish	Variable
P9GT32016G	Petri dish	Variable
P9GT20016G	Petri dish	Variable

I-2. 10-gal Drum #2

The total weight of 10-gal Drum #2 was 34 lb, net weight 16.5 lb, net volume 5 gal, <0.5 mR/hr @ contact. The drum contents are 17 interstitial soil squat jar samples, three Series 743 sludge squat jar samples, and five Series 741/742 sludge squat jar samples.

Table I-2. Drum identification (#2) and associated sample identity contents and approximate amount of sample for each drum repackaged for future retrieval purposes.

Glovebox Excavator Method Sample Repackaging		
Drum Identification: 10-gal Drum #2		JAN 24 05 GEM Squat Jar
Sample Identity	Sample Container	Approx. Amount
P9GT33016G	Squat jar	300g
P9GT36016G	Squat jar	300g
P9GT31016G	Squat jar	300g
P9GT35016G	Squat jar	300g
P9GT20016G	Squat jar	300g
P9GT28016G	Squat jar	300g
P9GT24016G	Squat jar	300g
P9GT30016G	Squat jar	300g
P9GT27016G	Squat jar	300g
P9GT23016G	Squat jar	300g
P9GT05016G	Squat jar	300g
P9GT25016G	Squat jar	300g
P9GT21016G	Squat jar	300g
P9GP02015G	Squat jar	300g
P9GT29016G	Squat jar	300g
P9GT22016G	Squat jar	300g
P9GP04015G	Squat jar	300g
P9GP01015G	Squat jar	300g
P9GR04012G	Squat jar	300g
P9GR23012G	Squat jar	300g
P9GR20012G	Squat jar	300g
P9GP03015G	Squat jar	300g
P9GT32016G	Squat jar	300g
P9GT26016G	Squat jar	300g
P9GT34016G	Squat jar	300g

I-3. 10-gal Drum #3

The total weight of 10-gal Drum # 3 is 36 lb, net weight 15.7 lb, net volume 5 gal, 0.5 mR/hr @ contact. The drum contents are 19 interstitial soil squat jar samples, 20 overburden soil puck samples, and five Series 741/742 sludge puck samples.

Table I-3. Drum identification (#3) and associated sample identity contents and approximate amount of sample for each drum repackaged for future retrieval purposes.

Glovebox Excavator Method Sample Repackaging		
Drum Identification: 10-gal Drum #3		JAN 24 05 #4, Squat Jar
Sample Identity	Sample Container	Approx. Amount
P9GT14016G	Squat jar	300g
P9GT03016G	Squat jar	300g
P9GT11016G	Squat jar	300g
P9GT17016G	Squat jar	300g
P9GT13016G	Squat jar	300g
P9GT10016G	Squat jar	300g
P9GT02016G	Squat jar	300g
P9GT16016G	Squat jar	300g
P9GT06016G	Squat jar	300g
P9GT12016G	Squat jar	300g
P9GT08016G	Squat jar	300g
P9GT15016G	Squat jar	300g
P9GT05016G	Squat jar	300g
P9GT19016G	Squat jar	300g
P9GT07016G	Squat jar	300g
P9GT09016G	Squat jar	300g
P9GT04016G	Squat jar	300g
P9GT01016G	Squat jar	300g
P9GT18016G	Squat jar	300g
P9GP03015G	Puck	40g
P9GP05015G	Puck	40g
P9GP01015G	Puck	40g
P9GP02015G	Puck	40g
P9GP04015G	Puck	40g

I-4. Summary of Isotope Amounts

Table I-4. Summary of isotope amounts listed in the three 10-gal drums (Drum #1, #2, and #3) that are placed in one 30-gal drum as determined by gamma spectroscopy.

10-gal Drum # 1		
Isotope	Ci	nCi/g
²⁴¹ Am	6.87E-04	2.08E+02
¹⁴⁴ Ce	9.91E-07	3.00E-01
¹³⁷ Cs	8.16E-10	2.47E-04
¹⁵² Eu	1.37E-08	4.16E-03
²³⁷ Np	6.52E-07	1.98E-01
²³⁹ Np	7.86E-09	2.38E-03
²³³ Pa	4.14E-09	1.25E-03
²³⁴ Pa	2.67E-09	8.08E-04
²³⁹ Pu	4.16E-03	1.26E+03
²⁴¹ Pu	8.75E-05	2.65E+01
²³⁷ U	1.76E-07	5.31E-02

10-gal Drum # 2		
Isotope	Ci	nCi/g
²⁴¹ Am	4.21E-03	5.62E+02
¹⁴⁴ Ce	4.37E-06	5.83E-01
¹³⁷ Cs	7.05E-09	9.40E-04
¹⁵² Eu	4.82E-08	6.42E-03
²³⁷ Np	2.36E-06	3.14E-01
²³⁹ Np	2.62E-08	3.49E-03
²³³ Pa	1.57E-08	2.10E-03
²³⁴ Pa	3.74E-07	4.99E-02
²³⁹ Pu	1.83E-02	2.44E+03
²⁴¹ Pu	2.02E-03	2.69E+02
²³⁷ U	4.91E-07	6.54E-02

Table I-4. (continued).

10-gal Drum # 3		
Isotope	Ci	nCi/g
^{241}Am	4.44E-04	6.25E+01
^{144}Ce	1.32E-07	1.86E-02
^{137}Cs	5.13E-10	7.22E-05
^{152}Eu	0.0E+00	0.0E+00
^{237}Np	0.0E+00	0.0E+00
^{239}Np	0.0E+00	0.0E+00
^{233}Pa	5.99E-09	8.43E-04
^{234}Pa	4.99E-08	7.02E-03
^{239}Pu	7.36E-04	1.04E+02
^{241}Pu	2.69E-04	3.79E+01
^{237}U	2.43E-07	3.42E-02

Table I-5. Summary of unvalidated isotope amounts listed in drum to be shipped for disposal through INTEC.

TRA050020		TRA050021	
Isotope	Curies	Isotope	Curies
²⁴¹ Am	1.71E-04	²⁴¹ Am	1.16E-06
²⁴³ Am	6.98E-10	²⁴³ Am	2.89E-10
²³⁷ Np	2.38E-06	²³⁷ Np	3.31E-11
²³⁹ Np	6.98E-10	²³⁹ Np	2.89E-10
²³³ Pa	5.43E-10	²³³ Pa	3.31E-11
²³⁴ Pa	6.23E-09	²³⁴ Pa	5.83E-11
²³⁸ Pu	8.09E-07	²³⁸ Pu	3.35E-07
²³⁹ Pu	4.73E-04	²³⁹ Pu	3.67E-06
²⁴⁰ Pu	5.19E-07	²⁴⁰ Pu	2.15E-07
²⁴¹ Pu	3.55E-04	²⁴¹ Pu	1.47E-04
²³⁵ U	8.33E-10	²³⁵ U	3.45E-10
^{108m} Ag	3.15E-11	^{108m} Ag	1.30E-11
^{110m} Ag	6.10E-12	^{110m} Ag	2.53E-12
¹⁴⁴ Ce	9.01E-08	¹⁴⁴ Ce	3.73E-08
⁶⁰ Co	1.82E-11	⁶⁰ Co	7.53E-12
¹³⁴ Cs	8.49E-13	¹³⁴ Cs	3.52E-13
¹³⁷ Cs	5.86E-10	¹³⁷ Cs	2.43E-10
¹⁵² Eu	4.54E-09	¹⁵² Eu	1.88E-09
¹⁵⁴ Eu	3.42E-10	¹⁵⁴ Eu	1.42E-10
¹⁵⁵ Eu	1.34E-10	¹⁵⁵ Eu	5.55E-11
⁵⁴ Mn	2.28E-11	⁵⁴ Mn	9.45E-12
¹²⁵ Sb	7.58E-11	¹²⁵ Sb	3.14E-11
²³⁴ U	4.15E-09	²³⁴ U	3.05E-10
²³⁷ U	2.06E-08	²³⁷ U	8.52E-09
²³⁸ U	7.42E-08	²³⁸ U	5.83E-11
⁶⁵ Zn	5.87E-12	⁶⁵ Zn	2.43E-12